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A review on electrospun membranes for potential air filtration application

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25 **Abstract**

26 Air pollution is one of the major environmental concerns in most highly populated cities,
27 which is typically caused by particulate ($PM_{2.5}$ and $PM_{0.1}$) or gaseous pollutants. In this
28 framework, membranes produced by the electrospinning technique are attracting more and
29 more interest thanks to their peculiar properties such as interconnected pore structure,
30 tunable porosity and fiber dimension, high surface area to volume ratio and controllable
31 morphology. This review aims to provide an exhaustive overview on the electrospun
32 membranes applied in air filtration introducing the key principles and fundamentals of the
33 separation mechanisms and discussing the influence of membrane properties (e.g.,
34 morphology and charge) on their filtration efficiency. The materials generally employed
35 for the fabrication of electrospun membranes (polymers, solvents) and their combination
36 with additives with defined properties are reviewed also in light of the new environmentally
37 friendly approaches which are increasingly adopted in membrane fabrication. Finally, the
38 practical use of electrospun membranes in several application fields such as individual
39 protection devices, environmental remediation, recovery of volatile organic compounds
40 (VOCs), and ventilation and climate control aspects is widely discussed providing also an
41 outlook on the upscaling potential of electrospun membranes and future directions.

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43 **Key Words:** Electrospinning; electrospun nanofiber membranes; air filtration; aerosols;
44 air pollution.

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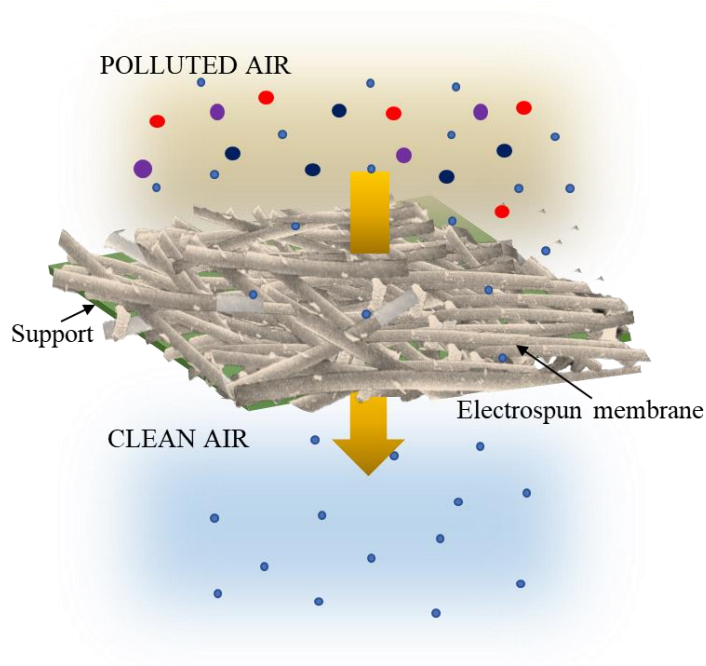
49 **1. Introduction**

50 The World Health Organization (WHO) has estimated and declared that around seven
51 million people die per year due to air pollution. Atmospheric pollution is a serious matter,
52 which can be classified into indoor and outdoor pollution. The first one, also named
53 household air pollution, is caused by burning, in inefficient stoves or open hearths, solid
54 fuel sources, including firewood and crop wastes, for cooking and heating. Among these
55 non-controlled human activities, a number of compounds are extensively produced such as
56 methane, carbon monoxide, particulate matter (PM), and volatile organic compounds,
57 resulting in a negative impact on human health. The exposure to these compounds becomes
58 relevant for low- and middle-income countries that often do not have access to clean fuels.
59 On the other hand, outdoor air pollution, or ambient air pollution, is primarily caused by
60 elemental necessities and activities in combustion processes from industries, motor
61 vehicles, power generation, agriculture/waste incineration, among others. This later type
62 of pollution is responsible for 4.2 million deaths per year due to heart disease, lung cancer,
63 stroke, and respiratory diseases [1]. Apart from the inherent pollution of human activities,
64 other natural sources of pollution are also produced such as pollen, spores, fungi, bacteria,
65 and virus carrying-aerosols that can be responsible for various health effects, including
66 asthma, allergy reactions, and infectious illnesses, such as influenza and chronic pulmonary
67 diseases [2].

68 The hazardousness and toxicity of inhaled particles strongly depends on their dimensions.
69 For instance, if thoracic particles with a diameter lower than 10 μm (PM_{10}) are partially
70 blocked by nasal cavities [3,4], fine (diameter lower than 2.5 μm ($\text{PM}_{2.5}$)) and ultrafine
71 particles (diameter lower than 0.1 μm ($\text{PM}_{0.1}$)) can be deeply inhaled into the lungs and be
72 accumulated on the alveoli causing a series of harmful effects [5] by crossing into the

73 pulmonary and systemic circulations and directly affecting the heart and blood vessels [6].
74 Therefore, there is a need of implementing devices with ability to sequestrate both natural
75 and anthropogenic **pollutants** from the air. In this sense, various air filtration materials have
76 been intentionally studied over the last years to protect human health against particle
77 pollution [7]. Potentially, membrane filtration can act as a physical barrier and thus remove,
78 depending on the membrane pore size, various types of particles, molecules, contaminants,
79 and microorganisms. Ideally, a membrane aimed for air purification must display the
80 ability to retain exclusively the pollutant particles while facilitating the transport of air [8].
81 In addition to this, membranes should also meet other requirements in terms of high
82 lifetime, low-pressure drop, easy to handle and installation, and low-production cost [9,10].
83 Regarding the particle's separation efficiency, current conventional air filtration
84 membranes are based on micro-sized fibers, which in fact are not efficient enough to
85 remove smaller contaminants than the range of 0.1-2.5 microns (i.e., PM_{0.1} and PM_{2.5})
86 due to their larger pore size [11]. At this point, electrospun membranes are becoming more
87 and more attractive as a key tool to satisfy such efficient air filtration performance due to
88 their interconnected pore structure, tuneable porosity, and fiber dimension (from 40 to 2000
89 nm in diameter), high surface area to volume ratio and controllable morphology [12,13].
90 Graphically, **Figure 1** describes a typical electrospun membrane that present a membrane
91 support.





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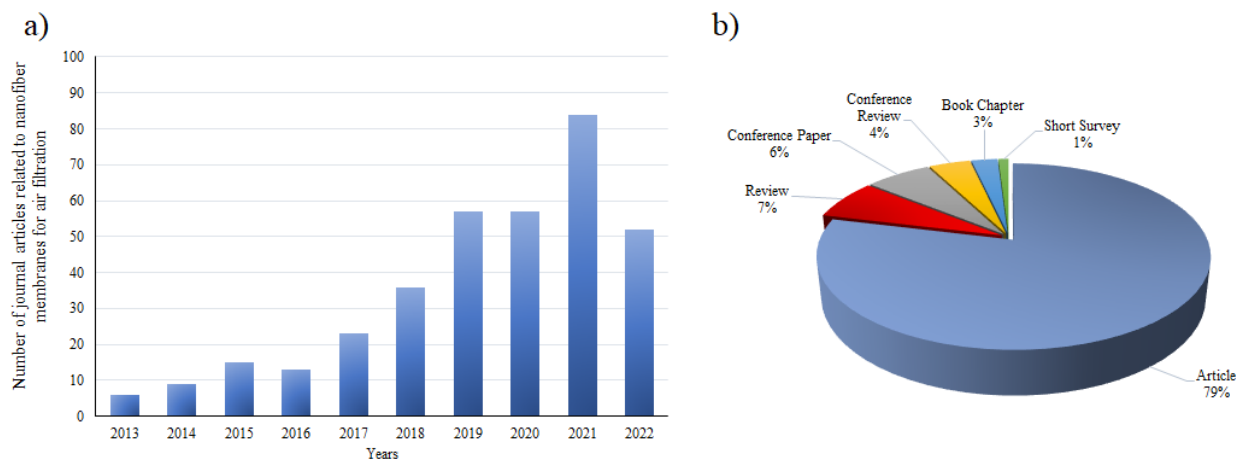
93 **Figure 1.** Graphical depiction of a typical electrospun membrane used for air filtration.

94

95 Electrospun nanofiber membranes are mostly fabricated by means of electrospinning
 96 technique, which is a very versatile technique allowing the tailoring of fibers with a very
 97 narrow diameter, a large specific surface area, a tuneable pore size, and more importantly,
 98 it gives the possibility to blend different types of nanoparticles (e.g. catalysts,
 99 antimicrobial agents) into the nanofibers and thus enhance their properties and purification
 100 efficiency [14,15]. Importantly, nanofibers can also be directly electrospun from a polymer
 101 dope solution as self-supporting membranes, but generally, the nanofibers are deposited
 102 onto a substrate made of polyethylene terephthalate (PET) or polypropylene (PP), which
 103 acts as a supporting layer conferring mechanical stability to the nanofiber.

104 **Figure 2.a** shows the publications during the last ten years. It is clear that the academic
 105 interest in this research area has continuously increased and by 2019 over 50 papers per
 106 year were published. **Figure 2.b** provides a general overview of the publications by type.

107 About 80 % of the publications used are articles published in academic journals. The other
108 20 % is divided by reviews, conference papers, book chapters, conferences and review
109 conference.



110

111 **Figure 2.** a) Journal articles and b) document types related to nanofibers for air filtration
112 over the past 10 years (updated until July 2022). Search engine: Scopus using the keywords
113 “nanofiber membranes” and “air filtration”.

114

115 Reviews on various aspects of electrospun membranes for air filtration have been published

116 in the last two years (2021-2022). Most of them focused on nanofiber materials for that

117 specific application. The research of Zhou Y. et al. [16], for example, based their approach

118 on the characteristics, advantages and disadvantages of single-polymer, composite or

119 hybrid nanofibers. Deng Y. et al. [17] proposed to assess the performance of nanofibers

120 prepared from bio-based polymers and novel bio-matrix such as cyclodextrin, lignin and

121 konjac glucomannan (KGM). Another interesting work was also proposed by the group of

122 Lu T. et al. [18]. They summarized the performance of electrospun air filtration membranes

123 (EAFMs) with different structures such as nanoprotrusion, wrinkled, porous, branched,

124 hollow, core-shell, ribbon, beaded, net structures. Ji et al. [19] introduced some basic

125 concepts involved in particulate matter (PM) filtration, such as classification and source of

126 PM, classic filtration mechanism, and the key evaluation parameters of PM filtration.
127 Moreover, Schneider R. et al. [20] described recent advances on the post-modification
128 strategies of spun micro/nanofibers surfaces using 0D, 1D, 2D, and 3D inorganic structures
129 while the group of Valecia-Osorio L.M. et al. [21] studied the electrospinning process for
130 air filtration nanofibers through three moments: solution preparation, fabrication
131 parameters and post-treatment techniques. In this review, we have provided a
132 comprehensive overview of the different aspects of electrospun membranes for air
133 filtration. We initially introduced the key principles and fundamentals of the filtration
134 mechanisms involved in electrospun nanofibers for air filtration, along with the influence
135 of membrane properties (e.g., morphology and charge) on their separation filtration
136 efficiency. Lately, we reviewed the preparation procedures employed for their fabrication
137 overlooking the membrane materials, solvents, additives, electrospun configurations. ~~and~~
138 ~~finally~~ Finally, we presented the practical applications of nanofibers in air filtration such
139 as individual protection devices, ventilation and climate control, recovery of volatile
140 organic compounds (VOCs) and environmental remediation. We also focused the attention
141 on the upscaling potential of nanofiber air filters, the current companies for electrospun
142 membranes production and the future challenges.

143

144 **2. Filtration mechanism in electrospun membranes**

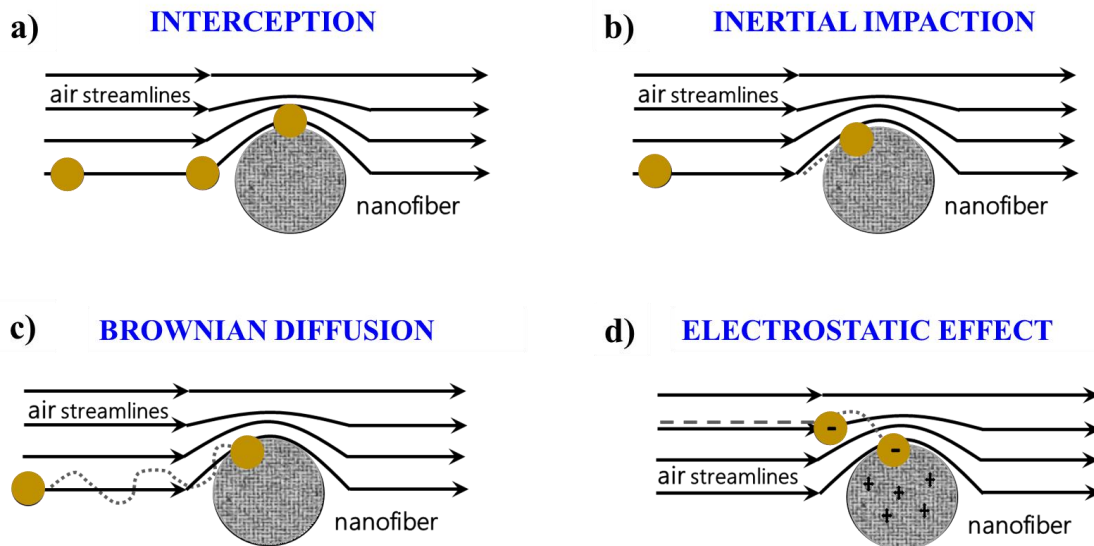
145 *2.1. Theory*

146 ~~Even if Air~~ filtration has a long history ~~and~~ consolidated over time and is closely linked to
147 industrial and technological developments. However, until the last century ~~that it was not~~
148 possible to find theoretical studies elucidating the various filtration mechanisms [22]. More

149 importantly, the lack of steady-state during filtration, which is characterized by the
150 variation of process efficiency over time together with airflow resistance because of
151 particles deposition, makes it challenging and limits a comprehensive description of the
152 phenomena [23].

153 Considering a moderate concentration of particles in the air, the particle accumulation on
154 the filter is minimal and does not alter the effective diameter of the nanofibers. Thanks to
155 this, the air filtration performance of an electrospun fibrous membrane is usually
156 considered stable (i.e. steady-state) [2]. Then, according to the widely known theory,
157 filtration occurs due to the following trapping mechanisms: interception, inertial impaction,
158 Brownian diffusion, electrostatic effect, and gravity effect, as represented in **Figure 32**.

159



160

161

162 **Figure 32.** Graphical depiction of the air filtration mechanisms through electrospun
163 nanofibers: a) interception, b) inertial impaction, c) Brownian diffusion, d) electrostatic
164 effect.

165

166 Typically, the particles tend to flow together with the air streamlines and are intercepted
167 while contacting nanofiber surface due to the van der Waals forces (see **Figure 3.2a**). This
168 latter phenomenon, so-called interception, is recognized as the primary filtration
169 mechanism. The interception is indeed independent of the flow velocity but still efficient
170 for capturing particles with size ranged from 0.1 to 1 μm [24].

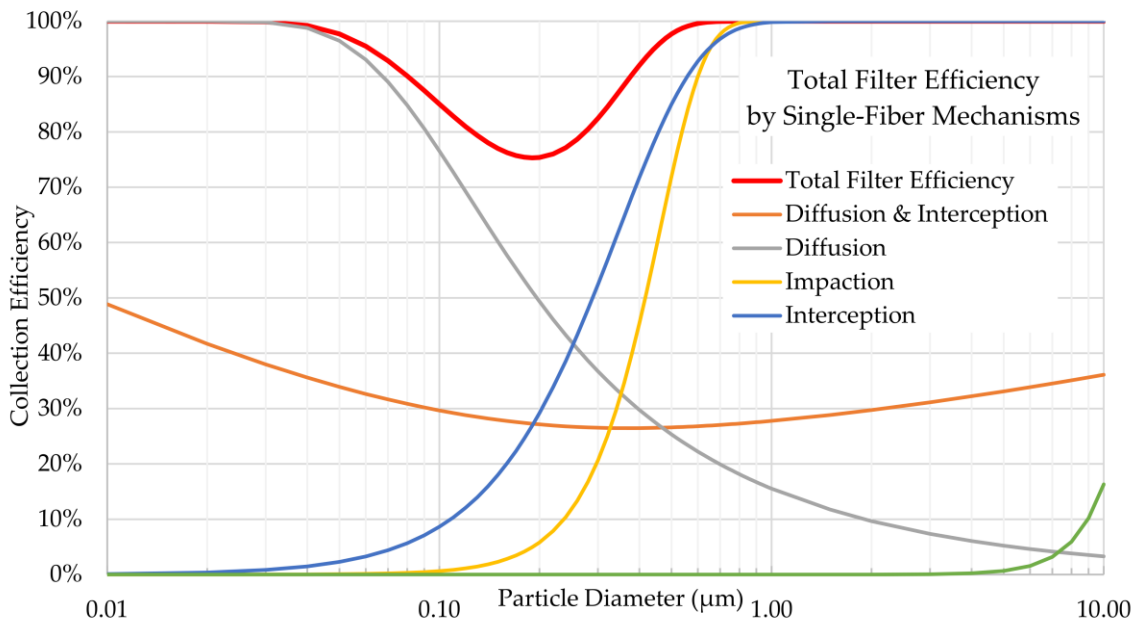
171 Since the electrospun membranes own randomly oriented nanofibers, the pathway of the
172 streamlines is commonly tortuous through air filtration. Particles often deviate from the
173 airflow line due to the effect of inertia force, impact subsequently against the nanofibers,
174 provoking an effective filtration thanks to the inertial impaction [25]. Of course, smaller
175 particles can succumb to the drag effect and be transported with the streamlines, while the
176 inertial momentum increases with the particle size and may overcome the drag force in the
177 case of larger nanoparticles, which causes a perceptible deviation from the streamlines and,
178 as a consequence, the impact against the nanofibers [26], as illustrated in **Figure**
179 **3.2b**. Herein, the efficiency of inertial impaction increases as a function of the particle size
180 and it is affected by the air velocity.

181 In addition to this, particles could slightly and arbitrarily deviate from their original flow
182 line thanks to the Brownian motion (see **Figure 3.2c**), and pronounced divergences
183 guarantying the diffusion of the particles towards nanofiber and then a consequent
184 interception [27]. This latter phenomenon typically occurs for particles smaller than 1 μm .

185 Besides the previous mechanisms, Coulombic attraction between charged particles and
186 unipolar or bipolar charged nanofibers substantially improves the filtering efficiency, as
187 represented in **Figure 3.2d**. It is worth mentioning that neutral particles can be also
188 polarized by unipolar or bipolar charged nanofibers or dielectrophoretic forces originated

189 by an external field [28]. Ultimately, the contribution of the gravity effect tends to be
190 negligible for particles smaller than 0.5 μm .

191 **Definitively, the sum of the contributions of each mechanism of filtration- which are**
192 **significantly affected by the particle size- determines the overall air filtration efficiency of**
193 **electrospun membrane (Figure 4).**



194 **Figure 4.** Filtration efficiency of a single nanofiber and contribution of the different
195 **mechanism of air filtration modelled with a Digital Twin [29]. This work is licensed under**
196 **a Creative Commons Attribution 4.0 International License.**
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199 2.2. Effect of membrane structure

200 As it is well known electrospinning is a versatile technique for the fabrication of efficient
201 air filters possessing high level of control on membrane morphology. Handling the
202 operative parameters of the electrospinning, as well as the chemical-physical properties of
203 the polymeric dope solution, is feasible to tailor different features of the nanofibers such
204 as their diameter, pore size, porosity and the packing density. These morphological and
205 structural features of the air filters meaningfully affect their performance. In general, the

206 filtration efficiency, represented as η , is usually described by the Kuwabara model, as
207 follows (1):

$$\eta = 1 - \exp \left[\frac{-4\eta_s \alpha L}{\pi d_f (1 - \alpha)} \right] \quad (1)$$

208

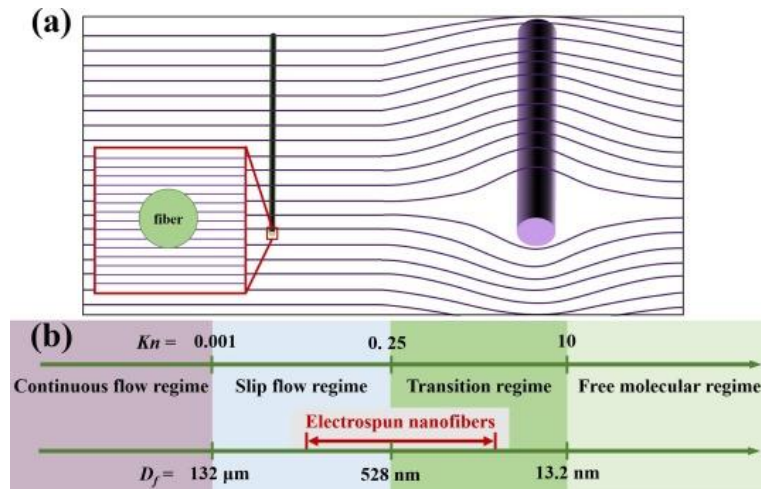
209 where η_s represents the filtration efficiency of a single fiber, α corresponds the fiber volume
210 fraction (i.e., packaging density), d_f represents the average fiber diameter, and L is the filter
211 thickness [30]. For instance, Eq. 1 spells out the robust dependence of the filter efficacy on
212 its structural and morphological properties, which is inversely proportional to the
213 nanofiber' diameter. Such description has been proven by experimental results as with
214 **polyacrylonitrile (PAN)** transparent electrospun nanofibers demonstrating a decrease of the
215 filtration efficiency (PM_{2.5}) from 98.11±1.41% to 48.21 ± 4.19%, as a result of their
216 diameter increase from ~0.2 to approximately 1 μm [31].

217 Surprisingly, nanofibers with a tiny diameter display minor pressure drop due to the slip
218 effect induced by unique aerodynamic conditions [32] (**Figure 5a**). Actually, the diameters
219 of electrospun nanofibers (d_f) and the mean free path of air molecules (λ) are comparable,
220 while the Knudsen number (Kn), expressed as $Kn = 2\lambda/d_f$, achieves values ranged from 0.1
221 to 10, which correspond to transition flow regime (**Figure 5b**). Here, the drag force is
222 substantially reduced and the air velocity on the nanofiber's surface is non-zero. This
223 means that the slip flow reduces the friction effect, the air molecules bypass the nanofibers
224 with the maximum probability of alleviating the pressure drop [31,33], as systematically
225 proved by a study on 122 nanofibers [34].

226 Of course, the high porosity is fundamental in reducing the pressure drop in combination
227 with “through” open pores, while “blind” closed pores remain as undesirable. Importantly,

228 pore size becomes also crucial for meeting a good performance in terms of filtration
 229 efficiency and permeability: small pore sizes gives an effective filtration but limit the
 230 permeability by promoting the pressure drop [35].

231



232

233 **Figure 5.** (a) The air flow field around two single fibers with different diameters according
 234 to the Kuwabara Model and (b) Knudsen numbers and fluid flow regimes of the electrospun
 235 fibers. Reprinted from [36] with the permission of Elsevier.

236

237

238 2.3. Effect of membrane and particle charge

239 As explained previously, **Brownian** motion is the random motion of particles suspended in
 240 the air, which is a result from their collision with the fast-moving of particles. However,
 241 this phenomenon is identified at a specific range of particle size. Particles with size smaller
 242 than $\approx 1 \mu\text{m}$ exhibit a significant Brownian motion [37], causing diffusion and deposition
 243 on the membrane surface, particularly in fibers [38]. Notably, Brownian motion becomes
 244 relevant with the decrement of particle size. Apart from described phenomena, if both
 245 particles/molecules or fibers are charged, the electrostatic effect is another parameter that
 246 interferes [39]. Theoretically, there are two ways to induce electrostatic forces in a

247 membrane air filter; the first one is directly related by charging the airborne particles, while
248 the second one can be a results of producing an electric field in the membrane [28]. In
249 recent times, this latter scenario has been investigated by embedding inorganic nanofillers
250 in membranes independently if the electrospun fibers already displayed any charge. It is
251 worth mentioning that within the electrospinning process, the used high positive voltage
252 may potentially confer a charge into the polymer solution, which certainly contributes to
253 the formation of volume charges and surface charges [7].

254 A charged particle (i.e. airborne particle) polarizes the fiber releasing a force that is equal
255 to the Coulombic force among the charge and the particle, but it is opposite to the charge
256 located inside the fiber at a position related to the optical image of the particle. Usually,
257 the image force is not highly crucial unless the particles own extremely high charges. The
258 electrostatic interactions, along with Coulombic interactions and polarization forces [40],
259 can indeed shift the pattern flow of particles in comparison with the initial air stream, this
260 may promote particles to deposit onto the membrane surfaces. Potentially, the electrostatic
261 effect will also promote the particle adhesion onto the fiber surfaces. This latter effect has
262 been commonly utilized in various aerosol filtration devices during the separation of sub-
263 micrometer particles [41], which in fact can enhance the particles' collection efficiency
264 [28]. As an example, nanofibrous mats containing boehmite nanoparticles, proposed as
265 electret filter, have shown exceptional separation performance by using the
266 nanomaterials, which played as an electrostatic charging agent [42]. In such research,
267 PA6 nanofibers were fabricated via electrospinning, obtaining an average fiber diameter
268 between 73-90 nm. Here, boehmite nanoparticles were smartly incorporated to induce
269 higher electrostatic charging characteristics of the unfilled PA6 nanofiber. It was concluded



270 that the filtration properties of PA6 nanofibers were mainly ascribed to the electrospinning
271 and the corona charging device [42]. Alternatively, the electrically charged nanoparticles
272 contained in polymer fibers may also provide another perspective when dealing with air
273 disinfection, since electrostatic interactions between microorganisms and charged particles
274 can lead to cell wall rupture, and thus the death of the possible bacteria, fungi, and viruses
275 [43,44]. This has been evidenced by Selvam and Nallathambi [45], who observed a 99%
276 bacterial filtration efficiency when using 10-12.4 wt.% of Ag nanoparticles into PAN
277 composite membranes. Ag nanoparticles have been also merged into electrospun fibers. In
278 this case, Zhang et al. [46] have utilized $-C(NH_2)=N-OH$ groups, coming from the reaction
279 of hydroxylamine and acrylic on PAN substrate, to smartly coordinate Ag^+ ions [46]. As
280 expected, the electrospun **nanofibers** displayed efficient antimicrobial properties, and no
281 substantial morphological changes were observed in the resulting fibers while maintaining
282 the filtration efficiency.

283 It is important to point out that electrically charged fibers tend to create in its vicinity an
284 electric field, which displays a force provoking the movement of a charged particle.
285 Apparently, the field produced by a charged fiber could polarize a particle as well,
286 unfortunately, such force on a polarized particle is considered negligible for small particles
287 [47]. In particular fibers, such as electret fibers, ~~they can display~~ a permanent line dipole
288 charge **can be observed which results** in a non-uniform field. Notably, the field may also
289 provoke a dipole charge in a particle but it does not represent a relevant **issue fact** for small
290 particles.

291 It is worth mentioning that the implementation of aerosol filtration via electrostatic forces
292 is likely to be challenging, but it has been already investigated. As an example, the Hansen



293 filter represents the first pioneering development and application of electrostatic forces in
294 aerosol filtration. Hansen discovered that the collection efficiency can be notably improved
295 when a wool filter was charged with colophony resin particles. Such an improvement
296 induces a better action of electrostatic forces [28]. Resin particles of approximately 1 mm
297 in diameter can be ~~be~~ negatively charged by contact (triboelectric charging). The charge
298 was maintained on the resin particle for a long time due to the fact that the resin owns good
299 electrical isolate agent. Electret filters have been initially based on dielectric fibers with a
300 quasi-permanent electrical charge. Unfortunately, membrane filter devices charged
301 electrostatically were fabricated using polymeric fibers including PP, PET, and nylon-6
302 (PA6). All fibers show relatively high electrical resistivity allowing them to be electrified
303 via corona charging, triboelectrification and induction. The applications of such
304 electrostatically charged filters can be extended to other relevant approaches, such as face
305 masks, building air conditioning systems [48], automobiles, air purifiers, to mention just a
306 few [42,49,50].

307 In current times, researchers are endeavouring on the design and development of innovative
308 electrically charged membrane materials to fabricate more efficient filtration systems. Al-
309 Attabi et al. [51], for instance, developed silica doped electrospun nanofiber membranes
310 with tuned surface roughness for potential aerosol air filtration, which revealed filtration
311 efficiencies as high as 95%. The research also pointed out that both surface charge and
312 electrostatic attraction of aerosol particles could also foster the air filtration performance
313 improvement of tetraethyl orthosilicate (TEOS)/PAN electrospun nanofibers.
314 Simultaneously, the filtration efficiency of polyvinylidene fluoride (PVDF) electret
315 nanofiber filters was also evidenced by Sun et al. [52], who studied the dielectrophoretic



316 effect toward neutrally charged nano- and sub-micron aerosols. Their size was ranged from
317 50 to 500 nm. Theoretically, the dielectrophoretic effect involves the induction of dipole
318 charges on neutrally charged aerosols as long as they are close to the charged nanofibers;
319 afterward, the polarized aerosols can be electrostatically caught by the charged fibers [53].
320 Apart from the stability for 24 h operation, the PVDF fiber filters displayed high filtration
321 efficiency and low-pressure drops. Punctually, the long-time operating effectiveness of the
322 electret multi-layer membrane filter was credited to the superhydrophobic properties and
323 exceptional electrical resistant properties of PVDF, leading to suitable electrostatic charge
324 stability and thus stable filtration performance [54,55].

325

326

327

328 **3. Electrospun nanofiber materials for air filtration**

329 Most of the research ~~has been focused~~ ~~emphasized~~ on the exploration of different classes
330 of polymers ~~with the view of~~ for tailoring nanofibers properties to be used in different ~~for~~
331 ~~various~~ applications, but particularly for air filtration at room and high temperatures. The
332 choice of utilizing ~~several relative~~ materials either polymeric or ~~biopolymeric~~, organic or
333 inorganic additives, and appropriate solvents, for the fabrication of fibers ~~aimed for air~~
334 ~~filters~~ becomes relevant from the fluid dynamics point of view, since the properties of a
335 polymeric solution in terms of viscosity, surface tension, and conductivity may ~~have~~
336 ~~display~~ a great impact on electrospinning process parameters [35]. Initially, polymers, as
337 organic materials, have been primarily explored for the fabrication of electrospun
338 nanofibers with ~~an~~ application in the air filtration. Polyimide (PA) [56,57],



339 ~~polyacrylonitrile (PAN)~~ [58], polyurethane (PU) [11], polysulfone (PS) [59], polyamide-
 340 56 [60] and polyamide-66 (PA 6,6) [61] are, for example, among the most utilized
 341 polymers in nanofibers preparation ~~as enlisted in Table 1. enlists some of these polymers~~
 342 ~~successfully used in nanofibers prepared by electrospinning aimed for air filtration~~
 343 ~~applications.~~ However, according to the current necessities in finding better materials, the
 344 merging of inorganic materials into polymer phases has conducted to the fabrication of
 345 mixed matrix nanofibers or composites. Today, the preparation of this latter type of
 346 membranes stands out as compelling strategy at enhancing the filtration efficiency of
 347 nanofibers while improving their chemical, mechanical and thermal stability together with
 348 enhanced antimicrobial properties [55,62]. Various nanomaterials, such as TiO₂ [63], SiO₂
 349 [64], ZnO [65], and Ag nanoparticles [63], have been successfully imbedded into polymer
 350 nanofibers.

351

352 **Table 1.** Polymer electrospun nanofibers fabricated via electrospinning. Adapted from
 353 [35].

354

Polymers	Polymer conc. (wt%)	Solvent	Electrospinning parameters			Reference
			Voltage (kV)	Flow rate (ml/min)	Distance (cm)	
PVA	10	Boric acid (BA)	15	0.5	15	[66]
	35.5	N,N-dimethylformamide (DMF)/acetic acid (AA)	24	0.5	16	[15]
PVA/ TiO ₂	-	Water	14	-	15	[67]
PA 6/6	9-14	Formic acid (FA)	20-50	0.25	11-16	[15]
PA 6	12	2,2,2-tri-fluoro ethanol (TFE)	8-20	0.3	15	[68]
PAN	15	DMF	19	0.8	20	[15]
	-	DMF	12.5-22.5	2	10-16	[69]
PAN/ SiO ₂	-	DMF	30	1.5	15	[62]
PU	-	DMF	10-30	0.6-1.2	20	[70]

355



356 The following subsections give an overview of the different materials applied in
357 electrospun nanofiber fabrication via electrospinning **technique**.

358

359 *3.1. Polymers*

360 **Polyamide (PA)**, PAN, and fluoropolymers such as PVDF and polytetrafluoroethylene
361 (PTFE), are among the most **used preferred** polymers by the research community due to
362 their inherent high chemical and thermal stability [71,72]. They also offer the advantage of
363 excellent processability for spinning and versatility to be adapted in various membrane
364 preparation protocols. For instance, PA is a widely investigated polymer for tailoring
365 filters thanks to its ability to be dissolved by different solvents, such as formic or acetic
366 acid, or dimethylformamide (DMF), to mention just a few. It also displays resistance to
367 water and thus humidity. It is biocompatible and presents a good mechanical resistance.
368 Promisingly, several researches documented the preparation of fibers from two types of
369 PA, named PA 6 (Nylon 6) and PA 6,6 (Nylon 6,6) [73,74]. These membranes were able
370 to perform ~~for~~ air filtration due to interesting features including their small diameter,
371 narrow diameter distribution, large surface area, and electrostatic charge [75]. A
372 comparative analysis among PA 6 and PA 6,6 fibers has been done by Matulevicius et al.
373 [76], who concluded that PA 6,6 fibers were likely to be the most performing ones. **In**
374 **particular, electrospun** fibers formulated with 8 wt.% of polymer concentration exhibited **a**
375 **filtration** efficiency **ranging** from 84% to 90% with a quality factor (QF) from 0.0486 to
376 0.0749 1/Pa.

377 PAN is yet another synthetic polymer with exceptional thermal resistance (degradation
378 above 300°C). The polymer is inert to plenty of organic solvents and acids and certainly



379 displays good processability. PAN fibers also represent a **valid** choice in producing carbon
380 membranes, e.g., they **can** be used as precursors for high-quality carbon fibers [77]. It has
381 been reported that PAN nanofibers have demonstrated high mechanical and thermal
382 stability, together with good efficiency for gaseous pollutants filtration in respect to other
383 polymer fibers [15]. Additionally, they can be operated in a wide and extreme hazardous
384 air-quality conditions (**PM_{2.5}** index >300, exhibiting an efficiency of 95-100% ascribed to
385 the surface capture ability and higher dipole moments [31].

386 Fluorine polymers on their own present also advantages **due to, such as** presence of the C-
387 F bond, which is stronger with respect to the C-H bond. The use of F instead of H in the
388 C-H bond fosters the bond strength that passes from 99.5 kcal/mol for the C-H bond to 116
389 kcal/mol for the C-F bond, resulting in a higher thermal and chemical stability of PTFE,
390 ~~this is because more~~ **because of the high** energy **is** needed to break the C-F bond [78]
391 **considering** that the fluorine atom is larger than hydrogen and it has unshared electron pairs
392 with higher electron density. Polymers presenting more fluorine atoms (such as PTFE) own
393 a high melting point, low coefficient of friction (e.g., PVDF: 0.3 Dynamic and PTFE: 0.04
394 Dynamic), and low surface tension (e.g., PVDF: 25 dyne/cm and PTFE: 18 dyne/cm).
395 Interestingly, the C-F bond is mainly responsible for the insolubility of the polymer in
396 common solvents; PTFE, for instance, is fully overlaid with fluorine atoms and frequently
397 demands high temperatures to be solubilized; while, PVDF, presenting the C-F bond and
398 the C-H bond in the structure, **shows** greater flexibility to be solubilized in different
399 solvents. Fiber filters based on fluoropolymers tend to show high filtration efficiency with
400 low-pressure drops and good filtration stability over time. On the other hand, PVDF filters
401 present high hydrophobic nature and a suitable electrical conductivity, allowing them to



402 display good charge stability and stable filtration performance [54,79]. PVDF filters were
403 combined with the typical air conditioning filters meshes based on polyester, PA, and nylon
404 for the air filtration of ~~PM_{2.5}~~ (PM with aerodynamic diameter $\leq 2.5 \mu\text{m}$) [80]. Also,
405 polyvinylpyrrolidone (PVP) and silver nitrate crystals were incorporated into the dope
406 solution to reduce defects or beads ~~while benefiting also of the for~~ antibacterial properties.
407 The scope of this study was devoted to outline the importance of the materials, such as
408 polymers, meshes, and the operating preparation parameters (such as voltage: 40, 45, 50,
409 55, 60 kV) on the final nanofiber's features and thus performance. By using polyester 80
410 mesh material, the QF turned to be the largest at 40 kV (QF: $23 \cdot 10^{-3}$), the smallest at 45 kV
411 (QF: $21 \cdot 10^{-3}$), ~~with~~ a filtration efficiency of 90%. Thanks to the properties of PVDF, the
412 recent development works rely on the production of novel electret fiber filters with high
413 strength, low air resistance, and outperforming for long-term aerosol filtration [52,81].
414 Similar to PVDF, PTFE fibers stand out as alternative candidates for air filtration thanks
415 to their high-temperature resistance, low-pressure drops, and high efficiency. The filtration
416 operation presenting PTFE filters take places at the surface of the fibers, which make them
417 suitable for a number of applications in high-efficiency particulate arresting (HEPA) and
418 ultralow penetration air (ULPA) in class cleanrooms. Very recently, Xu et al. [66]
419 fabricated and then tested PTFE nanofiber membranes for fine particulate filtration. In this
420 study, the filtration performance against aerosol particles was as high as 98% with a
421 relatively low-pressure drop (ca. 90 Pa). At this point, the area of the filter acted as the
422 contact surface, and due to its microporous structure, expressed as millions of pores per
423 square centimetre, the submicron particles were successfully separated. It is important to
424 mention that the layer below played merely as a support and its contribution to the filtration



425 process was considered as negligible. Strictly, the evaluation performance of these fibers
426 must be suitable for the filtration of particle sizes comparable with dust particles, which
427 are much larger than the pore size of the fibers, permitting only gas molecules to go
428 through. Its main disadvantage deals with the fouling phenomenon that can occur at the
429 surface. In other words, the particles can potentially form a cake layer on the surface of the
430 material, which will definitely compromise its separation performance [82]. Aiming to
431 address this latter point, several cleaning strategies have been investigated to restore the
432 functionality of the membranes including the passive cleaning with carbon particles [66]
433 or synergic effects based on adsorption photochemical catalysis, non-thermal plasma
434 photocatalysis, among others [22].

435

436 *3.2. Biopolymers*

437 In recent times, there is a strong necessity to produce and apply biopolymers [83,84],
438 produced from raw materials, for manufacturing nanofibers aimed for different membrane
439 separation techniques, but specially for air filtration. As an example, polyurethane (PU) is
440 a polyester that can be found in soft and rigid forms. It has a hydrophilic nature and ~~it is of~~
441 ~~course~~ derived from renewable sources [85,86]. PU fibers have shown fast absorption of
442 volatile organic compounds (VOCs) at both surface and the polymeric matrix, revealing a
443 competitive uptake capacity compared with traditional adsorbents such as activated carbon
444 [70].

445 PVA is yet another biopolymer that owns a semi-crystalline structure presenting hydroxyl
446 groups and hydrogen bonds. Interestingly, it is a water-soluble polymer with exceptional
447 hydrophilicity and good adhesive and barrier properties [87]. It is worth mentioning that



448 has been investigated in producing membranes for selective gas and solvent separations
449 [71,88,89]. When dealing with air filtration applications, Lv et al. [90] blended PVA with
450 a high molecular weight polysaccharide, like konjac glucomannan (KGM), ~~to obtain~~
451 displaying an efficiency of 99.9% in air filtration testing. A mixture of DEHS and neutral
452 monodispersed NaCl were employed as model aerosol particles with diameter ranging from
453 300 nm to 10 μm .

454 Poly(lactic acid) (PLA), made up of elements similar to lactic acid, is characterized by
455 excellent processability and high melting point temperature between 170-180 $^{\circ}\text{C}$, and T_g
456 values in the range of 50 - 65 $^{\circ}\text{C}$ [83,91]. An important environmental aspect behind PLA
457 regards its a low carbon footprint , making it as a good candidate for the production of
458 fibers for air filtration [35,86,92]. Finally, poly(acrylic acid) (PAA) [93] polyethylene
459 oxide (PEO), polyvinyl acetate (PVAc) [15], keratin, chitosan [94], and aliphatic
460 polyhydroxy-butyrates (PHB) [95–97] are among other biopolymers to be potentially used
461 for nanofibers production and implemented in air filtration.

462

463

464 3.3. Nanomaterials and other additives

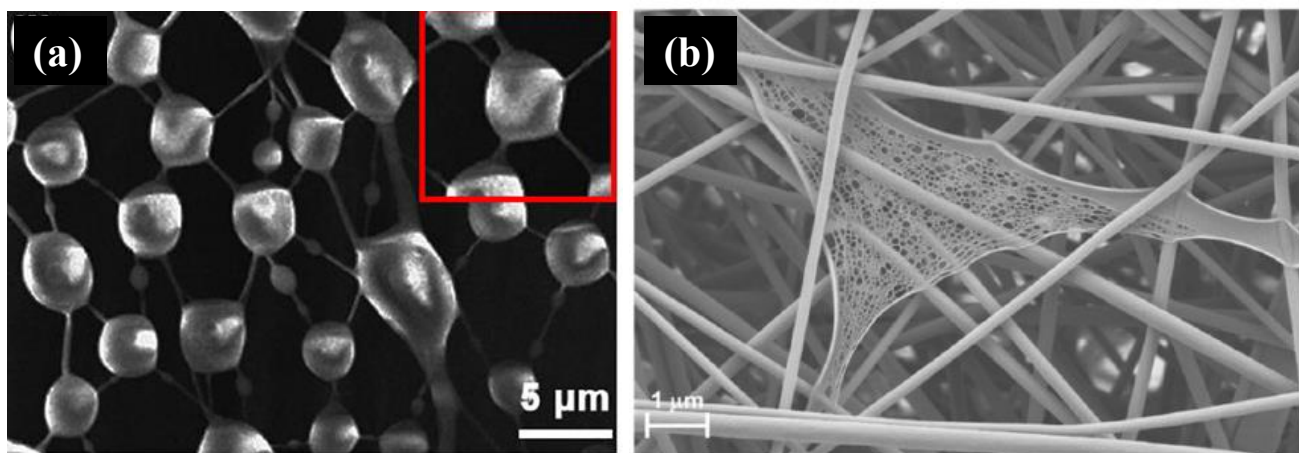
465 The application of additives, such as inorganic materials or salts, within the fabrication of
466 nanofibers for air filtration applications, are frequently needed to improve not only the
467 filtration efficiency but also the pressure drop and QF. Presently, it is a common practice
468 the blending of additives in the electrospinning technique in order to favour the formation
469 of nanofibers. For instance, the incorporation of additives results in a viscosity increase of
470 the solution and, in some particular cases, in the reduction of the fibers diameter of the



471 ~~fibers~~. Common salts are generally LiCl [79,98], tetraethylammonium bromide (TEAB)
472 [99], NaNO₃, NaCl, CaCl₂ [100]. ~~In the case of~~ LiCl has an electrical conductivity
473 estimated about 394 μS cm⁻¹ and it can be employed for changing the conductivity of the
474 solution and thus improving the charge density on the surface of the charged jet. This latter
475 fact becomes relevant in producing nanofibers with a more uniform morphology. TEAB,
476 for example, exhibits similar conductivity than LiCl in water. In general, salts dissolve in
477 the aprotic solvents in which the solvation can depend on the dielectric constant of the
478 solvent (known as general solvation), or the chemical structure of both solute and solvent
479 (known as specific solvation) [99,101]. Metal oxides, including Al₂O₃, TiO₂, SiO₂, β-
480 cyclodextrins (β-CDs) [55,67,102,103], and ZnO [90], are other additive agents that can
481 increase the surface roughness of the membranes, and particularly TiO₂ exhibits
482 antimicrobial and photocatalytic activities [104]. TiO₂ particles have been involved in
483 nanofiber production reducing the diameter of PS [59] and PVA nanofibers [105]. Another
484 important material is graphene oxide (GO), which has received considerable attention
485 lately [106–108]. GO has improved the filtration efficiency of PM_{2.5} (~ 99%) - **thanks to**
486 **its ability to effectively absorb PM_{2.5} particles (Figure 6.a)** - demonstrating an excellent
487 air filtration system in combination with PAN nanofibers [107].
488 Silver nanoparticles (Ag NPs), which are also known by their inhibitory activity towards
489 various bacteria [109], are typical dispersing inorganic phases in biocomposite nanofiber
490 fabrication. Zinc oxide nanoparticles (ZnO NPs) can improve the surface roughness
491 **resulting into** ~~translated to~~ a uniform narrow pore size distribution with an almost complete
492 air filtration efficiency (approximately 99.99%). β-CDs, which are cyclic oligosaccharide
493 presenting seven glucose units, are commercially available, low-cost, non-toxic,



494 biodegradable items derived from starch digestion. Such compounds have been recently
495 investigated in air filtration applications according to their particular cone-shaped cavity
496 structure. β -CDs have been involved for composite nanofibers preparation based on classic
497 polymers, such as PAN, where it has improved the adsorption capacity against VOCs.
498 Additionally, the combination of β -CDs with biopolymers, such as gelatin, allows the
499 production of bio-nanofibers ~~as a green solution and~~ as a green alternative solution to ~~the~~
500 ~~use of~~ fossil-based materials. As an example, composite gelatin/ β -cyclodextrin nanofibers
501 displayed the ability to separate aerosol particles (0.3–5 μm) with <95% filtration
502 efficiency at 0.029/Pa [110]. Such biodegradable nanofibers owned a uniform morphology
503 with a second “web” structure, characterized by superimposed and interconnected fibers
504 which presented a much smaller dimension than the main nanofibers, as showed in **Figure**
505 **6.b.**
506



507 **Figure 6.** (a) Absorbed PM_{2.5} particles on the PAN/GO. Reprinted (adapted) with
508 permission from [107]. Copyright 2019 American Chemical Society. (b) SEM images of
509 composite gelatin/ β -cyclodextrin nanofibers with “web” structure. Reprinted from [110]
510 with the permission of Elsevier.

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512
513

3.4. 4. Solvents for electrospun membranes

514 ~~The~~ Solvent is an important component ~~dealing with the crucial items needed~~ necessary
515 for the fabrication of most polymeric membranes ~~in membrane fabrication~~. This becomes
516 ~~relevant since it~~ In electrospinning processes, the solvent favours the formation of various
517 interactions between materials ~~in the electrospinning process~~. Also, the physicochemical
518 properties of the solvent can influence the morphology and more importantly the pore size
519 diameters of the resulting fibers. The solvent influences drastically the dope solution
520 properties such as viscosity and conductivity. To date, chloroform, DMF, and
521 dimethylacetamide (DMA) were found to be the best solvents for fabricating nanofibers.
522 Herein, solubility parameters are **essentially** pivotal to estimate solvent-polymer ~~the~~
523 affinity [111]. In theory, very close solvent-polymer parameters reveal a good solubility
524 and also the possibility to electrospin the solution at room or mild temperatures. With the
525 aim of tailoring defect-free nanofiber membranes, a second volatile solvent, such as
526 acetone or alcohols, are usually utilized in combination with the main solvent. The usage
527 of this **co-solvent** increases the time of solvent evaporation during the fibers formation
528 thanks to the high vapour pressure, ~~this promotes~~ **promoting** the production of uniform 3D
529 **nanofiber networks** ~~in nanofibers~~ [112]. National and international regulations limit the use
530 of dangerous chemicals and **encourage** ~~promote~~ the transition to more sustainable and less
531 toxic alternatives [113]. A series of other green solvents derived from biomass have been
532 recently proposed for the fabrication of polymeric membranes (in flat-sheet and hollow
533 fiber configuration) including dimethyl isosorbide (DMI) [114], γ -valerolactone (GVL)
534 [115], dihydrolevoglucosenone (Cyrene™) [116] ionic liquids [117] and/or deep eutectic
535 solvents (DESs) [118,119]. Recently, Russo et al. [79] produced PVDF electrospun
536 membranes using dimethyl sulfoxide (DMSO) as a low toxicity solvent in a mixture with



537 acetone. The results obtained have opened new perspectives for more sustainable
 538 production of electrospun membranes. The research in this direction remains open and
 539 attractive for the next future.

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549 **4. 5. Characterizations and performance evaluation**

550 It is widely known that the design and development of a new membrane concepts implies
 551 the characterization and performance when implemented in a specific membrane process.
 552 For instance, **Table 2** lists main **characterization** techniques used for the characterization
 553 of electrospun nanofiber membranes.

554 **Table 2.** Typical characterization methods and techniques used for electrospun nanofiber
 555 membranes. Adapted from [120].

Characterization	Method
Pore size	Bubble point method
Pore size distribution	Permporometry
Pore size distribution	Gas and liquid displacement methods (GLDP-LLDP)
Pore size distribution	Mercury porosimetry (MP)
Morphology, thickness	Scanning Electron Microscopy (SEM), Transmission Electron Microscopy (TEM)
Surface porosity, roughness	Atomic force microscopy (AFM)



Studies of chemical group	Infrared Spectroscopy (FT-IR, ATR, Photoacoustic)
Surface studies, chemical analysis	Energy Dispersive X-rays Spectroscopy (EDX or EDS), wavelength dispersive X-ray spectroscopy (WDS)
Thermal analysis	Differential Scanning Calorimetry (DSC)
Wettability	Contact angle measurement
Mechanical properties	Mechanical resistance, Stress-Strain measurements
Chemical stability	Swelling and contact angle

556

557 In general, the characterizations methods reveal important information about the
 558 morphology, structural, physical, chemical and thermal properties. ~~While~~ ~~When~~ dealing
 559 with air filtration, a dust filtration set-up is usually employed to evaluate the typical
 560 filtration parameters including filtration efficiency and pressure drop. In this experimental
 561 testing, particles, with size ranged from 0.6 to 180 μm , are commonly used and fed at a
 562 given air velocity [121]. The filtration efficiency of each fiber membrane ~~for air filtration,~~
 563 which results in overall filter efficiency, is ~~expressed denoted~~ as follows (2) (3) [39]:

564

$$\eta_s = \frac{\text{particles collected by fiber}}{\text{particles in a volume of air geometrically swept out by fiber}} \quad (2)$$

565

566

$$\eta = 1 - \exp(-\eta_s S) \quad (3)$$

567

568 where η_s refers to the filtration efficiency of a single fiber, S refers to the filter area factor,
 569 which is the projected area of fiber per unit of filter area, and η denotes the overall filter
 570 efficiency.

571 In addition to the high filtration efficiencies, electrospun membranes also display
 572 antibacterial properties during the filtration of air, which not only presents submicron

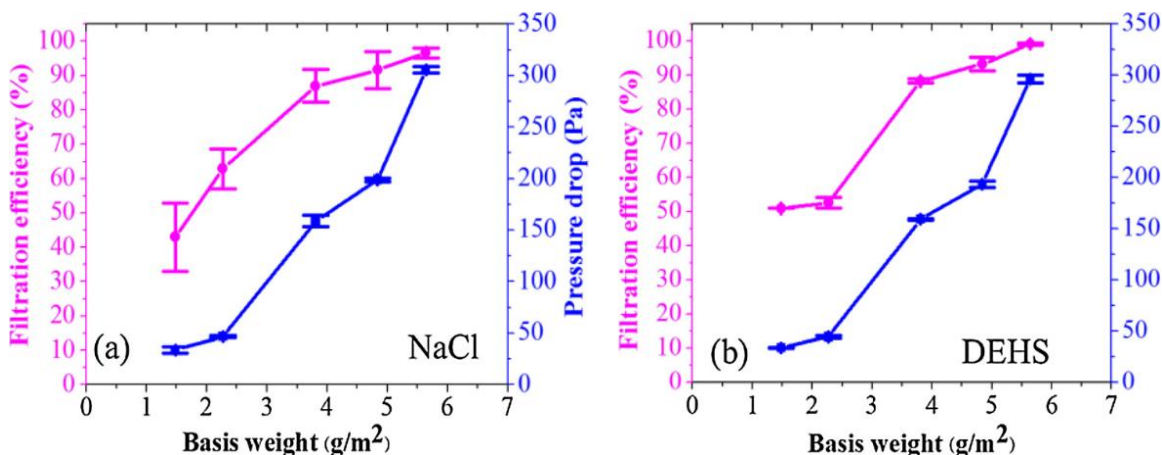
573 aerosol particles but also specific microorganisms (e.g., bacteria). Recently, Vanangamudi
574 et al. [55] engineered new composite nanofibrous based on PVDF and Ag–Al₂O₃, with
575 excellent antibacterial efficiency as high as 99.5%. Additionally, detoxifying properties
576 higher than 35% against paraoxon were observed. In aerosol filtration containing particles
577 (diameter of 0.36 μm), the filtration efficiency rate increased proportionally to the Al₂O₃
578 content, ranging from 2 to 8 wt.%. The filtration efficiency depended on the nanoparticle
579 loading, achieving ca. 99% filtration efficiency (at 8 wt.% Al₂O₃ loading). Furthermore,
580 the composite nanofibrous membranes revealed apparently stable antibacterial properties
581 over 99% towards *E. coli* [55]. In a different investigation, Ding et al. also evidenced
582 competitive filtration efficiency ranging from 80 to 90% of electrospun PVDF/SiO₂
583 nanofibrous membranes [122]. This latter development confirms the effect of nanoparticles
584 into pristine polymer nanofiber ~~with an~~ in enhancing the filtration rate. Concurrently,
585 Cannalli et al. [63], for instance, evaluated various inorganic particles into PAN nanofibers,
586 including TiO₂, ZnO, and Ag. Basically, all membranes performed with an average
587 filtration efficiency, but interestingly TiO₂-containing membranes owned the smallest fiber
588 diameter along with a complete filtration efficiency (ca.100%).

589 Shao et al. [123] have intentionally designed self-powered electrospun nanofiber
590 membranes for highly efficient air filtration. In this approach, the proposed electrospun
591 membranes were fabricated based on polyvinyl chloride (PVC) nanofibers and polyamide-
592 6 (PA6) nanofibers. By inducing air vibration, the triboelectric effect between both
593 adjacent nanofibers resulted in electrostatic charges, which contributed to observe
594 electrostatic adsorption capacity. Experimentally, the removal efficiency of the designed
595 nanofibers was as high as 98%, while the pressure drop was estimated as 67.5 Pa, this

596 proved a higher quality factor compared with the membrane lacking of electrostatic
597 charges. This development works proposed by Shao et al. [123] opens a new window **in**
598 **the fabrication of** outperforming membranes by exploiting the polymer features and the
599 electrospinning technique.

600 Zhu et al. [12] utilized silica nanoparticles with superhydrophobic nature which were
601 subsequently filled into a chitosan/poly (vinyl alcohol) (PVA) air filtration membrane via
602 green electrospinning and UV-cured nanofibers [12]. The inorganic hydrophobic fillers
603 were favourably added to form a rough surface and thus increase filtration efficiency. As
604 for filtration of fine particles (between 300 and 500 nm), the filtration efficiency and
605 pressure drop were noted to increase by increasing the basis weights, in other words, the
606 increase of the basis weight in membranes from 1.48 to 6.2 g/m² resulted in an
607 enhancement of the filtration efficiency from 42.97 % (with a pressure drop of 33.6) to
608 96.60 % (with a pressure drop of 305.6) for NaCl particles (**Figure 7.a**). Additionally, these
609 membranes showed a filtration separation towards di-ethyl-hexyl-sebacate (DEHS)
610 particles from 51% (with a pressure drop of 33) to 99 % (with a pressure drop of 296)
611 (**Figure 7.b**). Based on authors' conclusions, this phenomenon was credited to the high
612 basis weight, which **allowed confer** to the membranes to display more contact points while
613 increasing tortuous airflow channels [12].





614

615 **Figure 76.** Filtration performance of silica nanoparticles filled into a chitosan/poly (vinyl
 616 alcohol) (PVA) nanofibrous membranes with different basis weight investigated by using
 617 NaCl and DEHS particles. Reprint from [12] with the permission of Elsevier.

618

619 **Table 3** enlists a few examples of nanofiber membranes aimed at separating different
 620 pollutants from air. In general, it can be seen that most of the nanofibers display a good
 621 performance towards environmental remediation and mask filtration which is
 622 currently an area of research related to coronavirus disease.

623

624 **Table 3.** Performance of various nanofiber membranes for air filtration.

Pollutants	Materials	Efficiency	Pressure drop	Quality Factor (QF)	Notes	Ref.
		(%)	(Pa)	(Pa ⁻¹)		
PM _{0.3}	CNFs/PVP	86.4	17	0.117	For environmental remediation	[124]
	HNTs@CS/PVA/NWF	96.8	143.9	0.0239	For environmental remediation	[125]
	PP	87.28	40	0.052	For environmental remediation	[126]
	PVA (nanofibers deposited on PP nonwoven)	99.1	78	-	For ideal surgical mask and dust mask	[127]
	PLA	82.156	-	-	For ideal mask filtration	[128]
PM _{2.5}	PLA/ artificially cultured diatom frustules (DFs)	99.99	-	-	For ideal mask filtration	[128]
	PVDF	98.16	30	0.120	For environmental remediation	[129]
	TPP/N6	99.06	253	0.018	For environmental remediation	[130]

	PAA@ZIF-8	99.6	146.3	0.034	For environmental remediation	[131]
	PVA (nanofibers deposited on PP nonwoven)	99.9	56		For ideal surgical mask and dust mask	[127]
	PU/SiO ₂	95.37	126	0.001	For multilayer face masks (0.5 wt% of SiO ₂)	[132]
	PTFE/PP@PTFE	94.96	8	0.0348	for applications in nanogenerators, wearable electronics, medical products and other fields	[133]
NaCl aerosol particles	PMIA/PAN	99	-	-	For air pollution	[134]
	PTFE/PVA/ boric acid (BA)	98	30	-	For air pollution	[66]
	PVDF	99.2	-	-	For ideal mask filtration for SARS - COVID virus	[135]
	PVA/sodium lignosulfonate (LS)	99.44	24.5	0.212	For ideal mask filtration for SARS - COVID virus	[126]
Murine hepatitis virus A59 (MHV-A59)	PVDF	97.1	-	-	For ideal mask filtration for SARS - COVID virus	[135]
Diocetyl phthalate(DOP) aerosolparticles (0.3 μm) (TSI 3160 test)	PES/PAN	99.54	133.9	-	For ideal mask filtration for SARS - COVID virus	[136]
Diethylhexyl sebacate (DEHS) aerosols	PTFE/PP@PTFE	95.39	-	0.0358	For applications in nanogenerators, wearable electronics, medical products and other fields (DEHS size 0.200–4.595 μm)	[133]

625

626

627 **5. 6. Electrospun nanofibers configurations**

628 The material properties and the operating parameters of the electrospinning technique

629 greatly determines the configuration of nanofibers. **The configuration** influences ~~in~~ the

630 construction of two-dimensional (2D) and three-dimensional (3D) networks of nanofibers

631 [137]. Experimentally, the features of these networks are systematically analysed with the

632 aid of different characterization techniques, such as porosimetry, which determines the

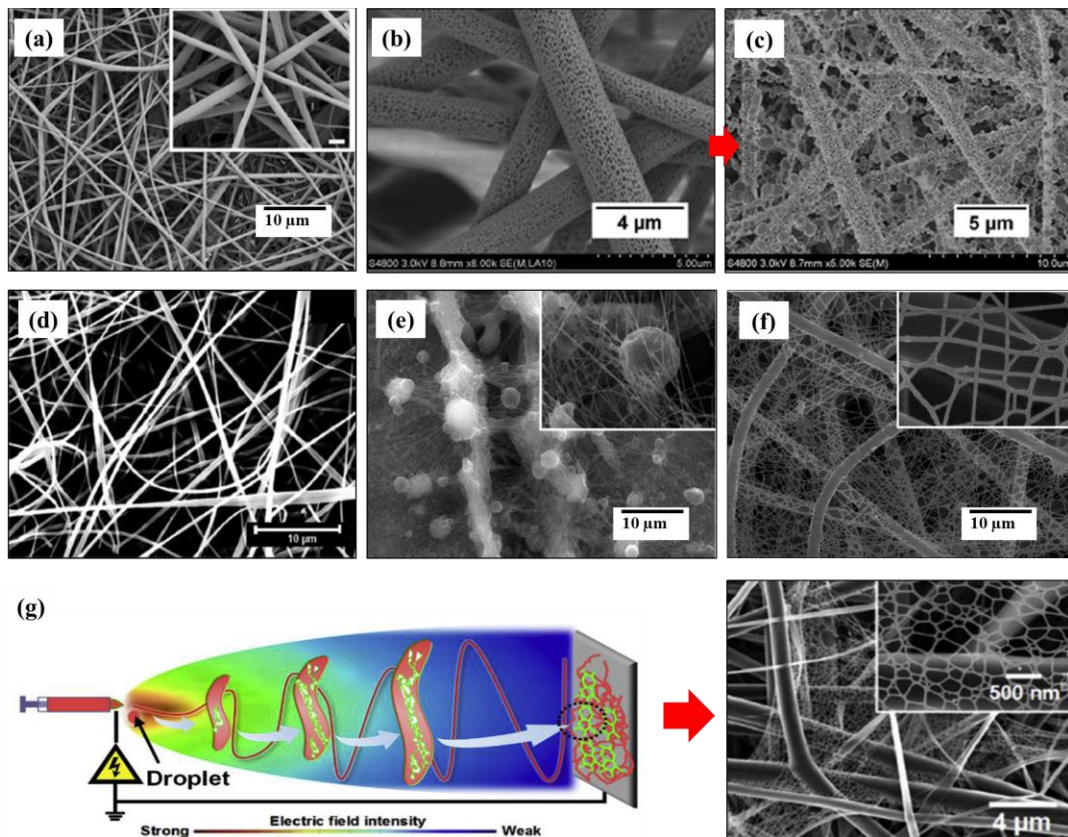
633 pore size, while scanning electron microscopy (SEM), transmission electron microscopy

634 (TEM), and atomic force microscopy (AFM) are used to investigate the morphology and

635 surface roughness. In the case of SEM, **this technique** is primarily applied for the
636 morphology study and thickness measurement of the nanofibers, while TEM tends to be
637 more convenient for the nanofibers having a diameter less than 300nm. The AFM is
638 commonly applied to characterize the total surface topography [138].

639 As for the pore size, polymeric nanofibers may present a pore size distribution ranged from
640 tens of nanometers to several micrometres; favourably, the submicron range is suggested
641 for better filtering efficiency. **Eventually,** A uniform and interconnected pore structure with
642 high surface area per unit volume is preferred as well [139].

643 The morphology of different polymer electrospun nanofibers based on PVDF, PAA, PAN,
644 PLA, CA is shown in **Figure 8**.



645

646

647 **Figure 8.** Morphology of different electrospun filtration membranes based on various
 648 polymers. (a) PLA, reprinted from [140] with permission of Elsevier; (b and c) Hierarchical
 649 structured nano-sized/porous poly(lactic acid) (PLA-N/PLA-P) composite nanofibers
 650 prepared at 45% RH of humidity before and after filtration test, reprinted from [141] with
 651 permission of Elsevier; (d) CA nanofibers with cationic surfactant cetylpyridinium
 652 bromide (CPB), reprinted from [142] with permission of Elsevier; (e) PVDF, reprinted
 653 from [143] open access; (f) PAN reprinted from [143] open access; (g) hypothetical
 654 situation simulation of electrospinning/netting and (h) nanofiber/nets structure of poly
 655 (acrylic acid) (PAA) with 0.03 wt% of NaCl, reprinted from [144] with permission of
 656 Elsevier.

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663 **6.7. Electrospun membrane applications for air filtration**

664 *6.7.1. Individual protection devices*

665 The current importance of electrospun membranes relies on the new perspectives for
 666 individual protection devices for air pollution or emerging infectious diseases (EIDs)
 667 including Coronavirus 2019 (COVID-19). A typical protector device for air filtration is
 668 characterized by fibrous (nonwoven filter) membranes composed by fine fibers. The
 669 mechanism of particles separation deals with an aerosol stream and entrapped between the
 670 fibers inside the filter [42]. **Table 5.3** provides a summary of regulations and/or tests used
 671 to certify masks elaborated by Das et al. [145].

672

673 **Table 5.3.** Regulations/test used to certify facemasks. Reprint from [145] with the
 674 permission of Elsevier.

Regulation/test	Scope	Limitation	Note
-----------------	-------	------------	------

Swedish Standard SS-EN 149 + A1:2009	Respiratory protective devices – Filtering half masks to protect against particles	At least 92% of the tested elements cannot leak N5% particles	Value taken for FFP3 device class.
NIOSH 42 CFR 84 US (NIOSH 1995)	Filtering capacity	At least 95% of the influent particles should be filtered	NaCl particles are used as surrogate particles. Typical values for N95 respirator masks.
T4D bacteriophage virus filtration, by [146] Relative survivability (RS) of MS2 viruses on filters, by [147]	Test the filtration property of different filter layers Test the ratio of virus survival on treated filters relative to untreated filters	1 h filtration time resulted on “infinite” T4D virus capture, while 2 h gave 1.1×10^8 1. PF PP filter (DuPont™ 01361 N): RS = 1 ± 0.1 . 2. CCF coarse pore cellulose filter paper (Whatman™ Grade 54): RS = 1 ± 0.2 . 3. FCF fine pore cellulose filter paper (Whatman™ Grade 50): RS = 1 ± 0.15	Based on a FFP2 mask
ASTM F2100 – 19e1	Standard specification for performance of materials used in medical facemasks	Medical facemask materials are required to: 1. Bacterial filtration N95% 2. Sub-micron particle filtration efficiency (0.1 μm) N 95. 3. Resistance to penetration of blood N80%. 4. Flame spread Class 1	Values based on a Level 1 barrier. 1. Bacterial filtration efficiency based on Test Method F2101. 2. Sub-micron Particle Filtration based on Test Method F2299 3. Resistance to Penetration by Synthetic Blood based on Test Method F1862. 4. Flammability based on 16 CFR Part 1610

675

676 The use of polymeric nanofibers is considered ~~recognized~~ as a new generation of filter
677 devices for enhancing the overall filtration performance in terms of efficiency and lifetime
678 compared with conventional systems. As previously mentioned, aerosol particles can
679 potentially be separated by different mechanisms including interception, inertial impaction,
680 gravitational settling, electrostatic attraction, and Brownian diffusion (dominant respect
681 others) [42]. The efficiency is dictated by the synergy of these mechanisms and it is
682 dependent on the particles size [148]. Certainly, other factors can also affect influence the

683 facemask efficiency including the facial fit of individual wearers and different mask types
684 commercially available [149]. As for HEPA masks, i.e., without the presence of
685 electrostatic attraction from the polymer fibers, an efficiency as high as 99.97% was
686 demonstrated at 0.3 microns [42]. It is well known that PP, PA, PAN, PVDF, and PTFE
687 are among the common polymers used for the production of facemasks. In a very recent
688 study, Shen et al. [150] developed PVDF/PTFE fiber layers and implemented them in the
689 facemasks, which were later evaluated for the removal efficiency of PM with different sizes
690 (from 0.3 μm to $>10 \mu\text{m}$). The outcomes demonstrated that for facemask wearers, the
691 exposure to ~~PM_{2.5-2.5}~~ concentrations in the air could be reduced by less than 20%. To
692 improve the electret performance and the uniformity of fiber structures, the perspectives
693 are oriented toward the design of novel network of nanofibers containing nanoparticles
694 [151], i.e., hybrid polymeric fibers [152]. However, the usage of nanoparticles may bring
695 another structural issues, for example, depending on the nanoparticles properties, a
696 potential agglomeration issue can be originated which can be translated to a non-efficient
697 separation performance [153].

698 Regarding the electric properties, these can be estimated by the electric intensity (\bar{E}) of the
699 fibers from surface potential (\bar{U}) analysis. The intensity can be calculated as follows (4):

700

$$\bar{E} = \frac{\lambda}{2 \pi \xi_0 L} \frac{\bar{U}}{\pi \phi (1-p) (\sqrt{R^2 + H^2} - R)L} \quad (4)$$

701

702 where λ corresponds to the charge density of a single fiber, ξ_0 refers to the dielectric
703 constant, L is the distance, ϕ and p correspond to the thickness and porosity of fibrous

704 membranes. R and H regard the radius of tested samples and distance, respectively
705 [152,154].

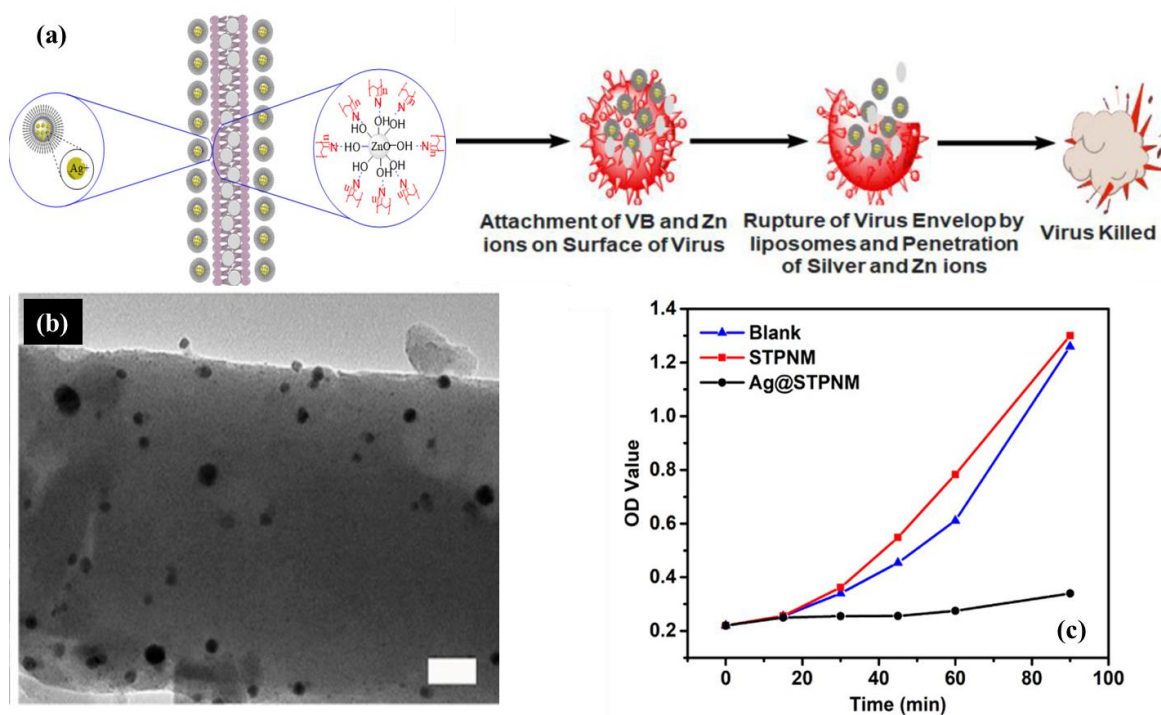
706 The \bar{E} parameter varies in an hybrid PS/PVDF nanofibers, and such a variation has been
707 investigated by Li et al. [152]. In general, such variation of \bar{E} parameter in the hybrid
708 PS/PVDF fibers was highlighted in a colour map for single PS and PVDF fibers. The
709 different surface potential of the materials was considered. As for PS, the red zone proved
710 the high electret effect on dielectric property ascribed to a positive surface potential (\bar{E}
711 positive); on the contrary, the graph for PVDF was characterized by negative \bar{E} values
712 credited to its negative surface potential, presumably for its polar nature.

713 During the development of anti-bacterial/antiviral materials for personal protective
714 equipment (PPE), photoactive chemicals were examined as effective for producing
715 antimicrobial electrospun membranes due to the possibility to produce oxidative biocidal
716 reactive oxygen species (ROS). TiO_2 , ZnO and Ag are considered as photoactive
717 nanoparticles used in the preparation of polymeric nanofibers that inactivate both Gram-
718 positive and Gram-negative bacteria [155,156]. Salam et al. [155] added the viroblock
719 (combination of silver and lipid vesicles) agent into a PAN/ZnO electrospun hybrid
720 membrane for a novel antiviral personal protective equipment (PPE). This viroblock is a
721 white viscous liquid that acts as an antiviral as well as an antibacterial agent. The
722 antibacterial efficiency for PAN/ZnO nanofibers loaded with 5% VB was 92.59% and
723 88.64% in the case of *Staphylococcus Aureus* and *Pseudomonas Aeruginosa*, respectively.
724 In a recent work, Zhang et al. [157] blended the Vitamin Ks (VKs) for producing antiviral
725 electrospun PAN membranes (VNFMs). The prepared VNFMs exhibited robust
726 photoactivity in generating reactive oxygen species (ROS) under both daylight (D65,



727 300–800 nm) and ultraviolet A (UVA,365 nm) irradiation, resulting in high antimicrobial
 728 and antiviral efficiency (>99.9%) within an exposure time of 90 min (**Figure 9.a**).

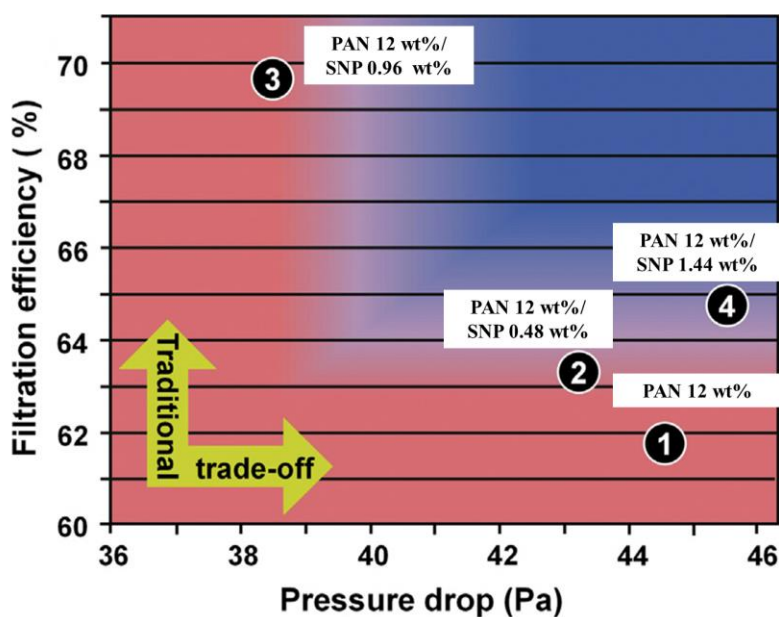
729 Wang et al. deeply described the benefits of the embodiment of SiO₂–TiO₂ porous
 730 nanofibrous membrane (STPNM) with Ag [158]. Interestingly, the inorganic nanofibers
 731 were prepared by electrospinning the precursors of SiO₂ and TiO₂ followed by a step of
 732 calcination, whereas Ag nanoparticles were embedded into the electrospun STPNM porous
 733 nanofibers through an impregnation process (**Figure 9.b**). The Ag@ STPNM electrospun
 734 membrane showed an efficiency of 98.84% in PM_{2.5} removal with a pressure drop of 59
 735 Pa coupled with an inhibition of the growth of *E. coli* of 95.8% (**Figure 9.c**).



736

737 **Figure 9.** a) Suggested mechanism of working of antiviral activity of VB-loaded PAN/ZnO
 738 electrospun nanofibers [155]. This work is licensed under a Creative Commons Attribution
 739 4.0 International License. b) TEM image and c) antibacterial performance of Ag@STPNM.
 740 Reprinted (adapted) with permission from [158]. Copyright 2019 American Chemical
 741 Society.
 742

743 Wang et al. [62] produced high-performance filtration membranes with structural
 744 alternatives by using different concentration of silica nanoparticles (SNP) into the
 745 electrospinning PAN solution. The presence of SNP on the filtration performance
 746 contributed to a high probability of particle capture due to the extended specific surface
 747 area and ~~can~~ acted as stagnant zones for airstream, generating low air resistance. The
 748 compromise between filtration efficiency and pressure drop was monitored by modifying
 749 the SNP content using the NaCl aerosol particles as reported in **Figure 10**.



750

751 **Figure 10.** Trade-off between filtration efficiency and pressure drop of relevant PAN/SNP
 752 electrospun membranes. Reprint from [62] with the permission of Elsevier.

753

754

755

756 *67.2. The role of electrospun nanofibers in environmental remediation*

757 The air pollution increment in highly populated cities **necessitates** the development and
 758 implementation of **new** efficient air **filtering systems**. Electrospun membranes, due to their
 759 outstanding properties, represent an **effective** alternative to be applied in these types of
 760 separation. Patanaik et al. [121], for instance, evaluated various types of PEO for the

761 synthesis of electrospun membranes, followed by their testing in air filtration applications.
762 Particularly, it was observed that by increasing the polymer concentration, from 3 to 6 w/v,
763 the diameter of the fibers increased from 85 to 125 nm, while improving their uniformity.
764 In air filtration testing, dust particles with size between 0.6-180 nm and fed at the constant
765 air velocity of 4 m/s, the membranes presenting the highest PEO concentration
766 demonstrated better performance in terms of dust particles retention of approximately 88%.
767 This latter outcome was ascribed to the fact that the polymer concentration increase
768 conducted to a reduction of membrane mean pore size (17 μm at 6 w/v of PEO) enabling
769 the membrane to retain more. Additionally, the membranes prepared at higher PEO
770 concentrations, resulting in a larger diameter, showed a lower pressure drop (17 Pa at 6
771 w/v of PEO) compared with the membranes having smaller diameter (28 Pa at 3 w/v of
772 PEO). In a different work, Canalli Bortolossi et al. [159] interestingly fabricated PAN
773 electrospun fibers containing particles at different concentrations (from 0 to 50 wt.%), and
774 deposited on a non-woven substrate. The main purpose was to design ~~a~~-nanofibers with the
775 ability to remove particles from the air. The final electrospun nanofibers owned a diameter
776 of approximately 250 nm except for the ones containing 10 wt.% of silver, displaying a
777 larger diameter (about 400 nm). This latter fact was basically ascribed to the change in
778 conductivity and solution viscosity ~~caused-provoked~~ by the embodiment of the silver
779 nanoparticles. Concerning to the air filtration test, the efficiency was initially analysed in
780 terms of pressure drop. The use of silver nanoparticles (up to 10 wt.%) resulted in an
781 increase in pressure drop (about 225 Pa) and consequently a decrease in the void space
782 which hindered the air permeation. Unfortunately, higher content of silver nanoparticles
783 (50 wt.%) resulted in a lower pressure drop (68 Pa). Such an explanation was justified

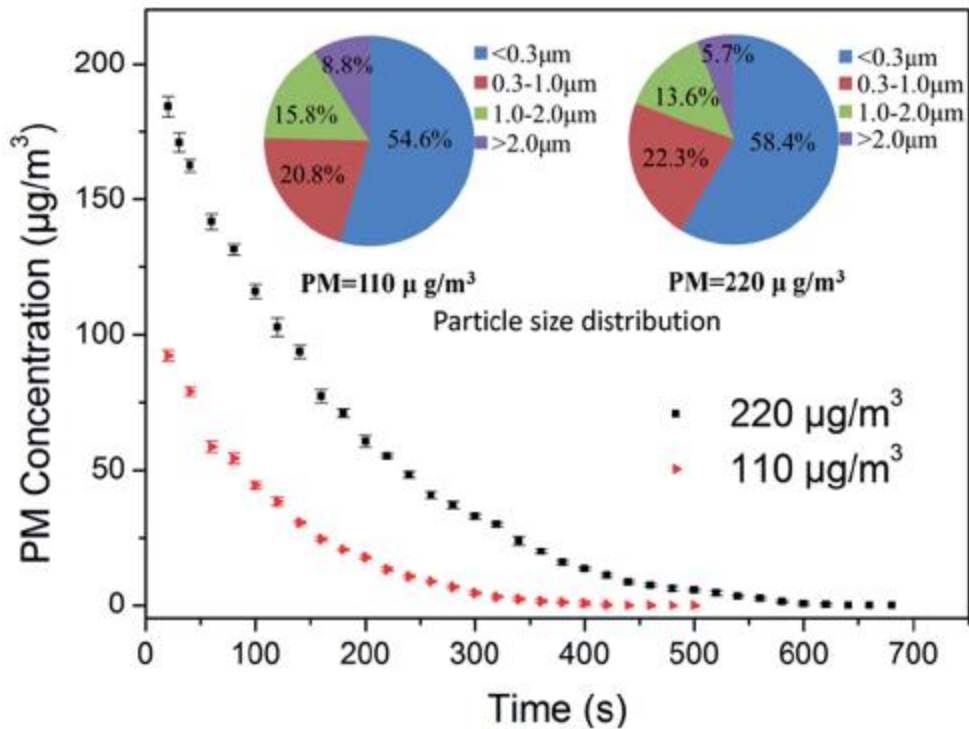


784 considering that the membranes containing the highest concentration of silver, thanks to
785 the higher solution viscosity, ~~were revealed a~~ **resulted in a** lower nanofibers deposition rate
786 on the collector. When the membrane yield was determined to separate nanoparticles
787 ranging from 9 to 300 nm, a complete removal, ca. 100%, was obtained for the samples
788 doped with a silver concentration up to 10 wt.% but, as mentioned previously, with a high-
789 pressure drop; however, a slight decrease in removal efficiency (approximately 98.6%) was
790 **observed seen** for the sample containing ~~with~~ the highest silver concentration (50 wt.%)
791 but with a much lower pressure drop as said above. Considering the obtained outcomes and
792 regulation established by the European Union Standard for EN 18222 filters, the membrane
793 lacking in silver loading could be categorized as H13, denominated as High-Efficiency
794 Particulate Air Filters - HEPA>99.95% collection efficiency. On the other hand, membrane
795 presenting 1 and 10 wt.% of silver, display the same performance as E12 (Efficiency
796 Particulate Air Filters – EPA > 99.5% collection efficiency), while membranes with 50
797 wt.% of silver meet the requirements for E11 filters (EPA > 95% collection efficiency).
798 Promisingly, credited to the presence of silver, the resulting silver/PAN nanofibers proved
799 also compelling antibacterial activity towards various *E. coli* bacterial strains.

800 The fabrication and subsequent implementation of polycarbonate (PC) electrospun
801 nanofibers for the removal of PM from the air was investigated by Li et al. [160]. In this
802 study, it is relevant mentioning that researchers conducted the air filtration experiments
803 using real polluted air (full of PM) collected from the outdoor campus of Zhengzhou
804 University (China). Unlike most investigations reported in literature where the air filtration
805 performance is determined using air models containing aerogel NaCl particles, which are
806 far to mimic, in terms of complex chemical composition, the PM dispersed in the polluted



807 air, in Li's study [160], researchers selected three levels of pollution varying in PM
 808 concentration: 50, 110, and 220 mg/m³. The filtration experiments notified how PC
 809 membranes were capable to separate and ~~this~~ retain the PM with an almost complete
 810 efficiency (approximately 100%), as reported in **Figure 11**. In addition to this, a good air
 811 permeability (78.36 ± 11.48 L/cm²h) was observed in such membranes. Taking into
 812 account that most of the PM owned a dimension lower than 0.3 μm, the membranes
 813 successfully operated in an outperforming way.



814

815 **Figure 11.** Filtration performance of PC membranes at different PM concentration and as
 816 a function of time. The particle size distribution is reported in the inserted pie charts.
 817 Reprinted from [160] with permission of Elsevier.

818

819

820

821 By comparing the filtration performance of PC membranes with PVA and polystyrene

822 membranes, it was concluded that the first ones were the most efficient to remove PM from

823 the air [160]. Favourably, it was found that the removal efficiency increased when the
824 polarity of polymers repeating units increased as well. Herein, it is quite possible that the
825 high dipole moment of PC membranes could have improved the binding of PM with
826 polymer nanofiber surface by means of dipole-dipole or induced-dipole interactions.

827 In a recent work, Cao et al. [161] prepared bead-free ~~polyacrylonitrile (PAN)~~-nanofibers
828 with diameter ~~lower than of~~ <100 nm to filter ~~PM_{2.5-2.5}~~-emissions from burning cigarettes
829 and ~~from~~ fused deposition modeling (FDM) three-dimensional (3D) printing. The
830 experiments ~~were~~ carried out by using two-chamber filtering devices. In the case of
831 filtration of cigarette smoke particles, the device consisted of two chambers as reported in
832 **Figure 12.a**. The dimension of smoke particles was in the range of 0.3-10 μm and the rate
833 of number emission was 3.9 x 10¹¹ particles/min and the ~~PM_{2.5-2.5}~~-emission rate was 2690
834 μg/min. In the case of particles filtration from FDM 3D printing, the device was prepared
835 by using an interior chamber with a hole covered by the layer of PAN nanofibers membrane
836 and an exterior chamber as reported in **Figure 12.b**. The emission rate of this type of
837 particles was fixed to 1-61 x 10¹⁰ particles/min. In these experiments, the efficiency was
838 calculated by using the equation (5):

$$\text{Filtration efficiency} = (1 - C/ C_0) \times 100\% \quad (5)$$

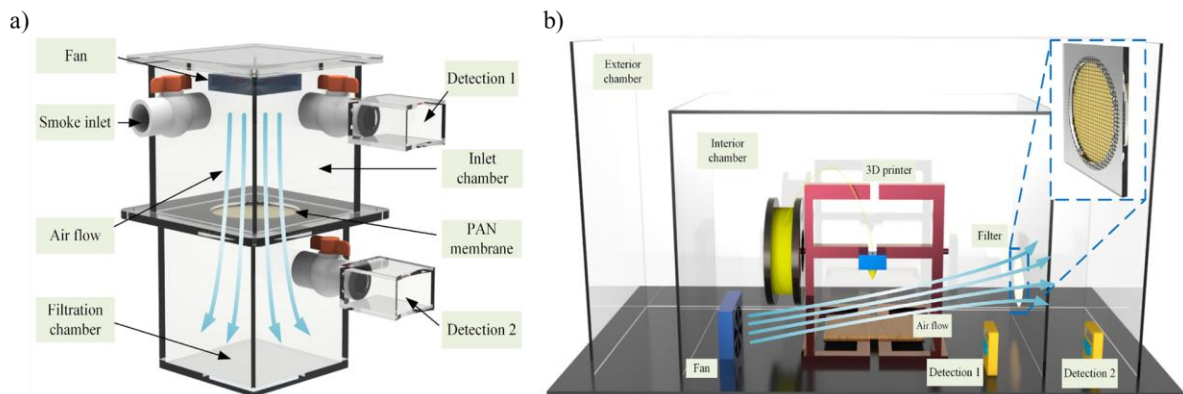
839

840 where C₀ is the PM concentration from Detection 1 (**Figure 12.a and 12.b**) and C is the
841 PM concentration from Detection 2 (**Figure 12.a and 12.b**).

842 The results confirmed a ~~an~~ ~~PM_{2.5-2.5}~~-removal efficiency of 99.26% in the cigarette smoke
843 particles filtration experiment and 81.16% in the case of particles sequestration experiment
844 from FDM 3D printing.



845



846

847 **Figure 12.** Set-up for a) cigarette smoke particles filtration experiment and b) particles
848 filtration experiment during 3D printing. Reprinted from [161] with the permission of
849 Elsevier.

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853

67.3. Electrospun nanofiber in recovering volatile organic compounds (VOCs)

854 The emissions of VOCs into the environment requires to be strictly regulated by legal limits

855 ~~since~~ due to their ~~negative~~ contribution to the production of polluted waste gas flows, and

856 to their harmful effects on plants, human beings and animals' health. Thereby, the control

857 and regulation of their emissions is a major worldwide concern. VOCs, methane, ethane,

858 tetrachloroethane, BTX, formaldehyde, acetaldehyde and acetone are among the most

859 common atmospheric pollutants, which are ~~the resulting emission of~~ various chemical and

860 petrochemical industries [162]. ~~Towards~~ For the recovery of VOCs, the techniques are

861 primarily categorized in two macro areas: process equipment modifications and add-on-

862 control techniques [162]. Within the second category, ~~we can find~~ it is possible to find

863 membrane ~~processes engineering~~ that represents an interesting alternative to separate

864 VOCs from gaseous streams. This is because membranes ~~processes~~ own several

865 advantages in terms of ~~the high chemical stability of specifically designed membranes~~ in

866 the presence of chlorinated compounds, together with the possibility to be operated at mild
867 temperatures, thus reducing the overall energy consumption [163]. The main property of
868 polymer membranes implemented used in membrane contactors for VOCs recovery deal
869 with is their high permeable-properties-permeation to VOCs while-substantially-hindering
870 the and low permeation to air [164]. Polydimethylsiloxane (PDMS) and polyimide (PI)
871 are typical membrane materials for the removal of VOCs, including acetone, toluene, and
872 xylene, from air or N₂. As-for PDMS, for instance, displays-a selectivity values ranging
873 from 11 to 25 in separating acetone [163,165], a selectivity of approximately 84 for toluene
874 removal and an CO₂/N₂ ideal selectivity of 21 [166]. Rebollar-Perez et al. [167]; for
875 instance, investigated the application of PDMS/ α -alumina membranes for vapor
876 permeation, constructing a device to remove VOCs from air currents (at feed pressure: 3
877 bar and temperature: 21°C). The preliminary outcomes proved that the membrane was able
878 to reduce the VOC content as high as 95%.

879 Scholten et al. [70] in their study fabricated and assayed electrospun polyurethane-based
880 fibers for the separation of VOCs from air, in which particularly was observed a complete
881 reversible VOCs absorption and desorption using a purge with N₂ at room temperature.
882 The final fibers exhibited a sorption capacity comparable to active carbon, which is
883 commonly used for vapor adsorption.

884

885 *67.4. Electrospun nanofiber in ventilation and climate control aspects*

886 Over the last two decades, membrane technology has attracted the attention in the field of
887 Heating, Ventilation and Air Conditioning (HVAC). From the environmental point of
888 view, the energy requirements of chillers and air conditioners are responsible for around



889 40–50% of the world's greenhouse gas emissions. Since the World Health Organization
890 declared the coronavirus disease 2019 (COVID-19) as a Public Health Emergency of
891 International Concern [168], it is highly needed to develop and implement new and highly
892 efficient filtration systems for air purification in closed spaces.

893 Vacuum membrane dehumidification, and membrane evaporative cooling and
894 humidification represent some of the HVAC membrane-aided processes, in which each
895 process operates differently in terms of inlets and outlets, together with different types of
896 membranes [169].

897 In the light of membrane-based air dehumidification process, the required membranes must
898 be dense with hydrophilic nature; requiring a driving force as the vapor pressure gradient
899 between the feed and permeate sides. In the case of membrane-based evaporative cooling
900 and membrane-based evaporative humidification process, they are categorized as low-cost
901 and energy-efficient technologies for the control of the evaporative cooling and the
902 humidity within the rooms, respectively [169].

903 Nanofibers based filters aimed for the filtration of small aerosols are usually produced by
904 electrospinning. Presently, Ju et al. [170] explored polyamide-6 polymer for the fabrication
905 of electrospun nanofibers containing silver nanoparticles. It is a common practice to
906 evaluate different operating parameters during the electrospinning. In this study, the
907 investigated parameters were the distance between needle and sample collector (18 cm),
908 processing temperature (40 °C), the voltage (18 kV), and the average flow rate (0.5 mL/h).

909 The aim of the research was also to evaluate their effect on the antibacterial and antiviral
910 properties of resulting air filter membranes for a high-efficiency PM removal. The
911 membranes displayed $PM_{2.5-2.5}$ -filtration efficiency as high as 99.99%, concurrently with



912 the removal of multiple aerosol pollutants and bacteria, such as *Escherichia Coli* and
913 *Staphylococcus Aureus*.

914 Photosensitized electrospun nanofibrous membranes were tailored by Shen et al. [135],
915 who aimed to capture and inactivate coronavirus aerosols. In this case, the rose bengal dye
916 was employed as a photosensitizer thanks to its exceptional reactivity in virucidal
917 generation. The electrospun membranes presented a pore size of ca.1.5 μm and a diameter
918 of approximately 200 nm. Finally, the filtration tests were performed using different virus
919 types, such as murine hepatitis virus MHV-A59, a coronavirus surrogate for SARS-CoV-
920 2. The findings evidenced a rapid inactivation of 98.9% after 15 min irradiation of
921 simulated reading light.

922 **8. Upscaling potential of nanofiber air filters and current companies for nanofiber** 923 **production**

924 Over the course of this review, we have reviewed one of the many applications of
925 electrospun nanofibers. To date, most of the research in electrospun membranes has been
926 done at a lab scale. Thanks to the outperforming separation of nanofibers for air filtration,
927 there is a current need of producing nanofibers on a large scale. However, the key
928 challenges in developing large-scale production of nanofiber rely on establishing accuracy
929 and reproductivity of the fabrication processing while satisfying the large volume
930 processing. Additionally, such processes must also meet important safety and eco-friendly
931 aspects of electrospinning. In this regard, centrifugal electrospinning [171], for instance,
932 owns the characteristics for large scale production at industrial level, along with high speed
933 and low cost [172]. It has been reported that such technique is able to tailor fibres with
934 diameters down to 100 nm [173]. Towards reaching scaling processes, higher flexibility



935 towards the materials and processing with multifunctional properties can be potentially
936 reached via co-axial and multi-axial technologies. Additionally, it has been documented
937 that ambient conditions drastically influence on the properties of electrified jets and
938 consequently on the resultant electrospun materials; to some extent, even small
939 environmental changes have demonstrated to have an effect on fiber features. Therefore,
940 several suppliers of commercial electrospinning devices have developed climate-controlled
941 electrospinning systems, assuring temperature and humidity control. For instance, IME
942 Technologies, recently named as Vivolta, <https://www.vivolta.com/>) fabricates laboratory-
943 scale systems for medical purposes, presenting electrospinning chambers and control of air
944 conditions, water filtration and automatization system.

945 In terms of industrial-scale equipment market of electrospinning devices, several
946 companies and suppliers have emerged satisfying this field, such as Elmarco
947 (www.elmarco.com), NaBond (www.electro-spinning.com), Holmarc Opto-Mechatronics
948 (www.holmarc.com), E-SpinNanotech (www.espinnanotech.com), Linari Engineering
949 (www.linaribiomedical.com), Kato Tech (www.keskato.co.jp), Mecc Co.
950 (www.mecc.co.jp), Toptec (www.toptec.co.kr), Electrospinz (www.electrospinz.co.nz),
951 Electrospunra (www.electrospunra.com), Vivolta Technologies
952 (<https://www.vivolta.com/>), Yflow (www.yflow.com), and Ino-venso
953 (www.inovenso.com). Importantly, electrospinning at industrial scale majorly implied a
954 rotating drum or on substrates using winding-unwinding systems, while laboratory set-ups
955 are based on needle-type electrospinning.

956 According to the experts in the field, to reach a fully implementation of electrospinning
957 systems at industrial scale, several aspects should be satisfied [173], as follows:

- 958 • As in most of emerging technologies, a substantial reduction in cost is highly
959 needed. This can be achieved as soon as potential stakeholder may be interested at
960 implemented electrospun nanofibers in commercial air filters.
- 961 • Multi-functional set-up should be fabricated, which is a current challenge since it
962 is difficult to offer “all-in-one” set-ups to reach different requirements from the
963 customer.
- 964 • Compactness in the devices is required since they need to be installed in limited
965 space.
- 966 • Productivity demand is also a current matter for large-scale electrospinning since the
967 nanofiber volume may be required by in many sectors.

968 When dealing with the industrialization and availability, various companies have released
969 electrospun products, such as Donaldson (www.donaldson.com), DuPont
970 (www.dupont.com), Ahlstrom Corporation (www.ahlstrom.com), Espin Technologies
971 (www.espintechnologies.com), Esfil Tehno AS (www.esfiltehno.ee), Finetex Technology
972 (www.finetextech.com), Hemcon Medical Technologies, Inc (www.hemcon.com),
973 Hollingsworth (www.hollingsworthvise.com), to mention just a few, in which the
974 nanofiber air-filter market has grown drastically [173].

975 To some extent, electrospinning devices should be also developed according to the needs
976 in terms of desired nanofiber properties, feedstock and items and, more importantly,
977 materials used for the fabrication of the nanofibers. In this latter aspect, new functional
978 materials are currently being developed to overcome the limitations of existing materials.
979 In this sense, bio-functional nanomaterials based on biomolecules, copolymers, and
980 polymer blends have been synthesized, such as fibroin in water-soluble polymers,

981 PCL/gelatin, poly(L-lactide acid)/gelatin blends, protein-based and chitosan-
982 poly(ethylene oxide)(PEO) blend [174–177], among others. Importantly, such materials
983 produced from natural sources have been proposed to face the eco-friendly weaknesses of
984 chemically synthesized polymers. Especially, natural protein nanofibers have shown high
985 efficiency filtration towards particulates, pollutants and toxic gases from polluted air [177].
986 However, typical polymers produced via chemical synthesized are still the preferred ones
987 for fabrication of electrospun mats for air filtration, as evidenced by Inovenso Ltd. Co.
988 [178]. It is worth mentioning that air filters presenting electrospun nanofibers are usually
989 applied in several applications, such as pulse-clean cartridges for Dust Collection.,
990 nanofiber filter media in Cabin Air Filtration of Mining Vehicles, and specialized face
991 mask fabrication, among many others [179]. All these applications will eminently foster
992 the establishment of electrospinning technology at industrial scale in near future.

993

994 **9. Grand challenges for electrospun membranes for air filtration**

995 The bottleneck of electrospun nanofibers is the technology transfer from the lab scale to
996 the mass production. In fact, the limited production rate of traditional electrospinning
997 equipped with a single needle has hindered the practical implementation of nanofiber for
998 air filtering at large scale (typically the flow rate of the polymeric solution is in the range
999 of $1\text{--}5\text{ mL}\cdot\text{h}^{-1}$ with a production rate of the nanofibers lower than $1\text{ g}\cdot\text{h}^{-1}$) [180].

1000 The employment of an auxiliar electrode could enable the ejection of up to 12 nanofibers
1001 from a single needle [181], but safety issues have limited the feasibility of this route to
1002 intensify the electrospinning process. As a manner of fact, the productivity of the
1003 electrospinning process increases as a function of Taylor cones. This has stimulated the

1004 development of multi-spinnerets where numerous needles are arranged in linear or two-
1005 dimensional arrays (es. circular, elliptic, hexagonal or triangular)[182] expanding the jet
1006 number up to 38,880 [183]. Moreover, an interesting advantages of multi-needle
1007 configurations is the opportunity to produce multi-component mat made of polymers not
1008 soluble in the same solvent [184].

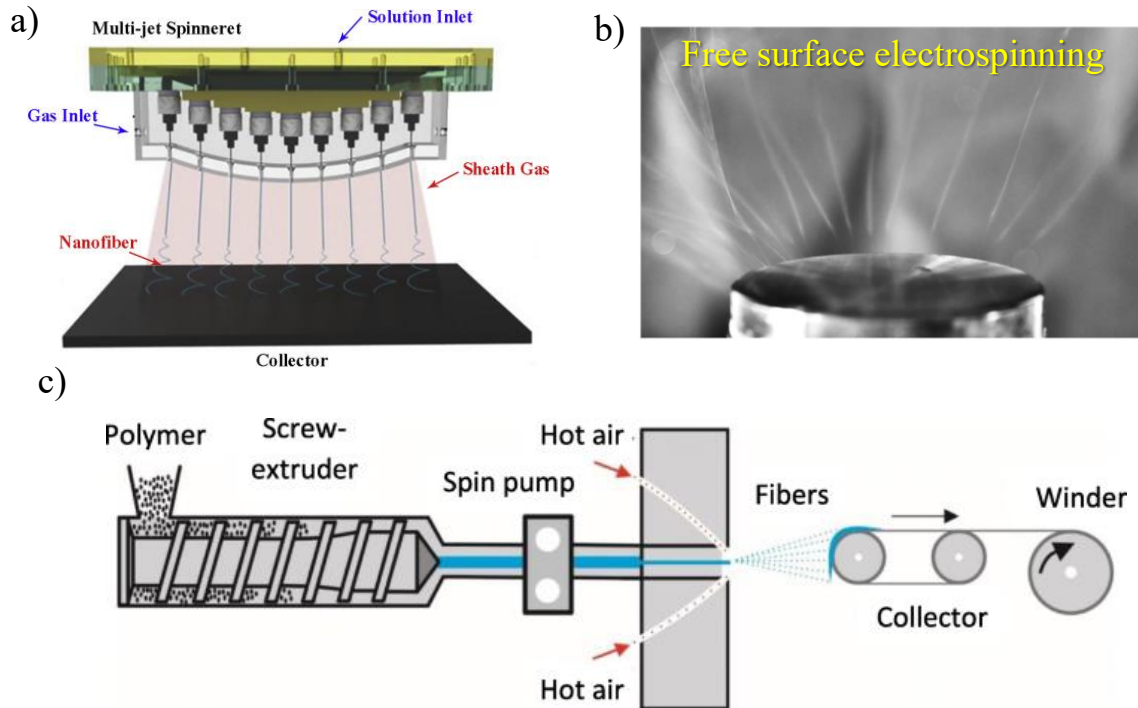
1009 Unfortunately, heterogeneous mat are often obtained with multi-needle spinnerets because
1010 of: i) the high density of needles which compromises the uniformity of the external field
1011 and ii) the Coulombic repulsions among the nascent fibers [185]. Several studies have been
1012 focused on the optimization of the multi-needle electrospinning focusing on the design
1013 (geometry, needle-to-needle distance, number of needles) of the spinnerets to: i) uniform
1014 the electric field, ii) minimize the interactions between the nascent fibers and iii) avoid
1015 their fusion during the flight from the needle to the support [186,187]. These critical issues
1016 have been also minimized by using polypropylene (PP) as dielectric material on the tip of
1017 the nozzles [188]. Another emerging and promising practice to mitigate the reciprocal
1018 interferences among the jets is the employment of a sheath gas in laminar regime (**Figure**
1019 **13.a**) which secured an improvement of the productivity of ca. 30-50 folds with respect to
1020 single needle configuration [184].

1021 Just to give an example about the scalability of the electrospinning of nanofiber with multi-
1022 needle spinneret, Inovenso Inc. has commercialized an industrial electrospinning system
1023 equipped with 110 needles able to produce up to 5 kg of nanofibers per day [189].

1024 Unfortunately, the improvement of the number of needles drastically increases the risk of
1025 clogging of the nozzles. A possible solution is to provide the polymeric solution to the
1026 outside surface of the needle. According to this strategy , thermoplastic polyurethane

1027 (TPU) has been electrospun with production rate of ca. $50 \text{ g}\cdot\text{h}^{-1}$ obtaining a network of
1028 nanofibers with a diameter of 145 nm and a $\text{PM}_{2.5}$ filtration efficiency of 99.9% [182].
1029 Recently, needless spinnerets able to eject nanofibers directly from the surface of the
1030 polymer solutions have been purposed (**Figure 13.b**). The mechanism of working of
1031 needless electrospinning-which is not influenced by the capillary effect- is based on the
1032 self-organization of the liquid solution at mesoscopic scale induced by electromagnetic
1033 field able to induce the expulsion of the nanofibers [190]. This strategy has the potential to
1034 ensure the industrialization of nanofibrous membranes for air filtering as demonstrated by
1035 technology developed by Elmarco Inc. using a wire electrode to eject multiple nanofibers
1036 which guaranteed an annual productivity of $20,000,000 \text{ m}^2$ ($0,03 \text{ g}\cdot\text{m}^{-2}$ of fibers of 150 nm
1037 of PA6)[191].

1038



1039

1040 **Figure 13.** a) Multi-jet electrospinning with sheath gas for the intensification of the
1041 nanofiber production. Reprinted from [192]. This work is licensed under a Creative

1042 Commons Attribution 4.0 International License. b) PAN nanofibers ejected from a self-
1043 made free surface electrospinning with a spherical section. Reprint from [193] with the
1044 permission of Elsevier. c) Scheme of a melt-electrospinning system. Reprinted from [194].
1045 This work is licensed under a Creative Commons Attribution 4.0 International License.
1046

1047 Another significant Achilles heel in the industrialization of nanofibers is the environmental
1048 impact. Tendentially, nanofibers are ejected from diluted polymeric solutions with a
1049 viscosity below than 20 poise (solvent content >70wt %) [138,195] and they are obtained
1050 upon the evaporation of the solvent during the flight from the needle to the support. Thus,
1051 mass production of electrospun nanofibers implies the use of enormous volumes of solvent
1052 raising serious concerns about the environmental footprint of nanofibers. Thus, it is
1053 necessary to introduce circular economy strategy to collect and recycle the solvent during
1054 the production of membranes via electrospinning. Noteworthy, common solvents used for
1055 electrospinning process are restricted by the Chemical Control Regulation in the European
1056 Union (REACH), such as halogenated (e.g. chloroform, trifluoroethanol) and toxic
1057 solvents (e.g. dimethylformamide) [196]. Beyond the employment of green solvent for the
1058 environmental-friendly production of nanofibers (see Section 3.4) , melt electrospinning
1059 (**Figure 13c**)- the process of spinning polymers from their melts- guarantees the solvent-
1060 free preparation of nanofibrous air filters [197]. Interestingly, melt electrospinning secure
1061 the preparation of i) nanofibers without residual of solvents [198] and ii) air filters of non-
1062 soluble polymers, such as polypropylene (PP) and polyethylene (PE) [199]. For instance,
1063 PP nanofibers with a diameter ranging from 7 μm to 14 μm were deposited on PP
1064 nonwoven support via melt-electrospinning. After post-treatment (i.e. hot-pressing), a mat
1065 of 0.42 mm of thickness showed a filtration efficiency above of 95% filtration for oil
1066 particles (size of 2.0 μm) and an air permeability of 54.69 $\text{mm}\cdot\text{s}^{-1}$ [200].

1067

1068 **7-10. Conclusions and future perspectives**

1069 ~~The growth of the human population, the increase of urbanization, and industrialization~~
1070 ~~have brought the decrement of air quality over the last few decades. Air pollution starts to~~
1071 ~~be a major issue that can result in serious risks for human health. In addition to this,~~
1072 ~~microorganisms and natural contaminants suspended in the air can potentially provoke a~~
1073 ~~great damage to human life. Looking for~~ Being considered efficient and reliable
1074 technologies, electrospun membranes are gaining a lot of interest in a plethora of
1075 applications in air filtration sectors ~~represent a viable tool and have been pointed out~~ for
1076 their ~~potential at enhancing~~ capacity to effectively improve the air quality ~~by~~ through a
1077 relatively simple filtration mechanism. Electrospun membranes ~~on their own~~ display
1078 ~~possess, in fact,~~ a number of advantages in terms of small **nanofibers** diameters, high active
1079 surface area, tuneable morphology, and interconnected pore structure. This review has
1080 provided an overview and discussed the potential trapping mechanisms which can occur in
1081 electrospun membrane filtration, as well as their effect on membrane separation
1082 performance. ~~We also reviewed~~ A number of applications reporting the successful
1083 ~~application use~~ of electrospun membranes for air filtration, and their comparison with
1084 commercial **filters**, was also reported ~~for such a purpose~~. The **removal** performance of
1085 electrospun membranes in removing different types of contaminants has been evidenced
1086 as high as 100% with a low-pressure drop up to 68 Pa.

1087 Although many developments and advances have been done in this field, many challenges
1088 still remain open and are waiting for potential solutions. Unfortunately, some of the
1089 bottlenecks of electrospun membranes comprise their poor mechanical stability which

1090 frequently demands their deposition on a suitable support, along with their compromised
1091 application under harsh conditions. An interesting finding in this review reveals that the
1092 reduction of fibers diameter certainly improves the filtration efficiency, but compromising
1093 unfavourably the mechanical properties.

1094 For wide exploitation of electrospun fiber, there is a need to ~~turn develop~~ the spinning
1095 technique into ~~an easy and affordable technology as easy as possible~~ with an applicability
1096 on a large scale [7]. Within the preparation processes, there is also an interest to replace
1097 traditional toxic solvents, that are typically used for the fabrication of electrospun
1098 membranes, ~~herein, the latent~~ with more benign alternatives, ~~are~~ the so-called green
1099 solvents [111,201]. This aspect becomes relevant when dealing with environmental
1100 protection and human safety. Some research groups ~~have already started~~ to explore new
1101 solvents to make the electrospinning process more sustainable and eco-friendly [79,202]
1102 outlining the importance of this aspect in developing the next generation of membranes
1103 aimed for environmental remediation.

1104 ~~As a recommendation for the new researchers in the field: great important~~ advances in
1105 nanofibers synthesis, either polymeric or composite, have been ~~done~~ over the last years;
1106 however, ~~they were limited to model tests on small lab scales. this review timely finds the~~
1107 ~~need to initiate the~~ The testing of electrospun membranes ~~for~~ with real air samples, as
1108 evidenced by Li et al. [160], ~~is, therefore, crucial. Most researches tend to use Air~~ model
1109 samples for performance evaluation ~~can reveal, in fact,~~ a good approximation but ~~a-~~more
1110 realistic outcomes, ~~on real case studies, will be determine the feasibility of new concepts~~
1111 ~~of nanofibers, fundamental to prove the efficiency of nanofiber-based membranes in air~~
1112 ~~filtration processes.~~

1113

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