Progress in Electrospun Nanofibres for Air Filtration

L R Manea¹, A P Bertea* and A Bertea¹

¹ Gheorghe Asachi Technical University of Iasi, Faculty of Industrial Design and Business Management, Prof. Dr. Doc. Dimitrie Mangeron Street, No. 29, RO-70005047, Iași, Romania

*E-mail: andrei_bertea@yahoo.co.uk

Abstract. Air pollution is a growing problem worldwide, reason for seeking more efficient methods to prevent exposure to contaminated air. Air filters must have the ability to effectively remove extremely small quantities of hazardous materials from the air, without causing excessive pressure drops, all at acceptable price conditions. A modern nanotechnology for obtaining polymeric nanofibres is electrospinning, capable of producing nanofibres of various materials, with controlled structures and porosity and with small pores, which makes it a favorable candidate for efficient air filtration. Electrospun nanofibres can be used directly or incorporated into various devices to reduce the negative impact of air pollution. The article briefly reviews the latest electrospinning technologies, highlighting the structural and performance advantages in producing air filters in what regards thickness, porosity, pressure drop, permeability antibacterial properties. The types of material used to obtain the nanofibres used as filtration medium are analyzed and the structural properties and performances of the filters are discussed. Finally, the challenges of electrospinning in producing air filters and future perspectives were summarized.

1. Introduction

Air pollutants are divided into two main categories: particulate matter (PM) and gaseous contaminants [1]. The air pollution is caused by the increase of the industrial emissions in the air and the exhaust gases of cars [2, 3], and has a growing impact on public health, as well as the environment, particularly fog and haze pollution [4-9]. The environmental impact refers to the influence of atmospheric particulate matter on Earth’s climate by scattering and absorbing solar radiation and by influencing cloud formation [10].

In terms of human health, it is known that mainly particles smaller than 2.5 μm in diameter, which cannot be blocked by the nasal cavity, have a strong negative effect on respiratory organs [11], as they penetrate into the lungs and cardiovascular system causing from irritation to chronic respiratory and lung cancer and worsening preexisting diseases [12, 13]. One of the most efficient methods to remove suspended particles from the air is air filtration, and electrospun nanofibre filters have proven to be amongst the most effective, due to their exceptional properties such as high specific surface area and high porosity with fine pores [14, 15].

2. Electrospinning principle

The 21st century began under the auspices of nanotechnology, with hopes of obtaining nano-materials with innovative properties and the exploitation of unique phenomena occurring at the nanometric scale [16]. All of these are expected to have a considerable impact in many fields, environmental protection
included [17]. As a result, nanotechnologies developed at the turn of the century are the foundation of future technological platforms [18].

Nanotechnologies aim to study and develop phenomena from nano-level materials in order to achieve effects and performances on a macro scale. The ability to manipulate individual molecules and perceive their characteristics at the atomic level has made this field very up-to-date.

Of all the technologies for obtaining nanofibres, the most efficient in terms of productivity and efficiency is electrospinning, a technology that evolved from electrospray technology [19]. This process is capable of producing long, continuous, controlled-oriented nanofibres with varying degrees of complexity and architectures, adaptable to different types of polymers. Electrospinning is identified as one of the key, leading technologies of the future [20, 21].

In the electrospinning process, to create an electrically charged jet of polymeric solution or melt, a high voltage current is used [22-27], respectively, one electrode is placed in the solution / melt, and the other is attached to the collector (figure 1). The electric field is applied to the capillary tube containing the polymeric fluid, maintained by its own surface tension. Thus, a charge is induced on the surface of the liquid and the mutual rejection of the electrical charges acts as a force directly opposite to the surface tension. When the intensity of the electric field increases, the hemispherical surface of the fluid at the tip of the capillary is elongated, forming a cone known as the Taylor cone. When the intensity of the electric field, in its growth, reaches a critical value, respectively when the electrostatic force of rejection exceeds the surface tension, the jet of electrically charged fluid is ejected from the tip of the Taylor cone. The jet thus formed begins a twisting process (in which the solvent evaporates, respectively in the case of melting, solidification takes place), leaving behind a polymer nanofibre with electric charge, which is randomly deposited on the collector.

Electrospinning results in a network made up of submicron fibres. Larger fibrous deposits can be obtained through the use of multiple capillaries. These fibrous deposits have many special properties, such as: low weight, high permeability and small pore size, which represent advantages in filtration applications, as well as numerous other areas of application such as: sensors, protective materials, drug controlled release, sound absorbers, composite materials, antimicrobial products, biological dressings, tissue repair, enzyme immobilization, etc [19, 21, 24, 28-36].
Electrospun fibre networks offer the advantage of advanced physico-mechanical properties. In addition, the amount of electrospun fibres required to obtain a product is low, and the electrospinning process itself is characterized by high versatility, allowing spinning of fibres with different sections, under various conditions, using a wide range of polymers [26, 27, 36 -38].

3. Applications of electrospun nanofibres in filtration

Nanofibrous materials are suitable for filtration applications, as a result of their special properties [11, 39]. Especially electrospun nano-fibrous filters showed high filtration efficiency, due to their exceptional properties in what concerns high specific surface area and high porosity [14, 15]. The use of the fibres obtained by electrospinning in the field of filtration covers two domains: air filtration, respectively water filtration (both in water and wastewater treatment).

The importance of using electrospun nanofibres in the field of air filtration is attested by the numerous patents [40]. In 2007 Ngiam observed that in the first decade of the new millennium 75% of existing patents in the field of nanofibres refer to the manufacture or use of nanofibres in filter systems and a decade later the patents in this field continued to be very numerous [41, 42]. The situation is similar in terms of scientific articles dedicated to electrospun nanofibres [43].

![Figure 2. Evolution of the number of papers dedicated to electrospinning and electrospun polymer nanofibres [43].](image)

Such a situation is normal if one takes into account the fact that conventional filters have certain flaws, particularly related to their efficiency in removing small particles from the air such as pathogenic agents like viruses. According to Barhate and Ramakrishna [44], high-efficiency classical filters are viable for particles up to 0.3 pm in size.

Atmospheric aerosols contain particles of different sizes ranging from ultrafine range (diameter less than 100 nm) to a gross range (over 2.5 pm); the size of viruses, for example, varies from 10 nm to 200 nm, bacteria from hundreds of nm to tens of pm, and pollen granules from tens to hundreds of pm. The fine particle range (less than 2.5 µm) can be divided into two categories: the core module (less than 40 nm) and the accumulation module (over 40 nm). Smoke and smog particles have diameters in these ranges [45, 46].

In order to protect from nanoparticles, which can have a detrimental effect on health, it is imperious necessary to increase the filter retention capacity and this can be achieved using electrospun nanofibre filters, which found use in domains as individual protection, building air filters and dust collection and vehicle cabin [47].
4. The main characteristics of air filters

The effectiveness of the filter materials is given by their capability to separate the aerosol particles from the air flow. In addition, the filtration efficiency must remain unchanged over time, under conditions of low airflow resistance and high loading capacity, without affecting the pressure drop. For this reason, the porosity of the filter materials used for this purpose is essential [48].

Air filters can be classified according to their ability to hold fine particles, respectively the smallest size of retained particles. According to the British classification system (ISO 16890) filters can be divided in 4 classes conformable to their efficiency: 1. ePM1 Filter efficiency for particle size range 0.3-1 µm; 2. ePM2.5 Filter efficiency for particle size range 0.3-2.5 µm; 3. ePM10 Filter efficiency for particle size range 0.3-10.0 µm, ISO coarse Gravimetric filter arrestance for filters not able to achieve minimum 50% efficiency at ePM10.

For high efficiency filtration HEPA (High-Efficiency Particulate Air) and ULPA (ultra low penetration air) filters are used [49]. These filters are usually made of glass and polyolefin fibres and show over 99.97% filtration efficiency for particles with a diameter greater than 300 nm [50].

Efficiency of fibrous filter commonly depends on the size of the particles, the airflow velocity and filter characteristics (fibre size and packing density). Filtration efficiency rates a filter by the percentage of contaminant removed as expressed in equation [51, 52]:

\[
\text{Filtration efficiency, } \% = 100 \times \left( \frac{C_1 - C_2}{C_1} \right)
\]

where \( C_1 \) = upstream particle concentration;
\( C_2 \) = downstream particle concentration.

The filter efficiency is expressed as a percentage (between 50% and 95% in 5% increases) for each of the ePM ranges indicated above.

Another important factor used to evaluate the filtration performance is the pressure drop, namely the differential pressure between upstream and downstream of the filter medium [51].

The aerosol particles are retained by filters by means of five mechanisms. The most predominant mechanisms are interception, inertial impact and diffusion [52-54], but gravitational deposition and electrostatic attraction can also occur [48].

\[\text{Figure 3. Mechanisms of particle capture [53].}\]

The prevailing filtration mechanism depends largely on the size of the particles, and the filtration efficiency is the result of all the present mechanisms. An estimated range of particles in which the aforementioned mechanisms manifest and the filtration efficiency depending on the particle size is shown in Figure 4.

It can be seen that diffusion is the prevailing mechanism for small particles having a diameter of less than 0.3 µm [48], similar to the electrostatic attraction, which only takes place when the particles and / or fibres have an electrical charge.
The particle size has a great importance on the filtering efficiency, as it can be seen from Figure 5, where uniform fibre with 5 μm in diameter, a packing density of 0.05 and a thickness of 2 mm have been used at an air flow velocity of 10 cm/s with 1 g/cm³ particle density.

![Figure 4. Main filtering mechanism (a) and filtration efficiency (b) depending on particle size [44].](image)

![Figure 5. Filter efficiency as a function of particle diameter [54].](image)

It can be seen that the filtration efficiency registers a minimum in the area of diameters between 300 and 700 nm, a zone that is called the most penetrating particle size (MPPS), and theoretical calculations forecast that the efficiency may be significantly increased using nanofibrous media [55].

The size of the electrospun nanofibres and the packing density can be controlled by varying the solution parameters (viscosity, conductivity and surface tension), ambient parameters (temperature and humidity) and the process parameters (fluid flow rate, spinneret to collector distance and electric field strength having a most significant influence on the properties of the obtained nanofibres) [56, 57], while the particle size as well as air flow rate depends on the application.

5. Characteristics of nanofibres that are recommended for use in air filtration

Conventional nonwoven fibre-based air filters have been used from many years for air filtration [35], as they are capable to effectively retain PM particles while letting air to pass through the filter, but these kind of filters frequently suffer from the drawbacks of too large pore size and low porosity due to their microsized diameters [58]. A solution to these disadvantages is the use of electrospun nanofibres based filters, which integrate the structural essential quality of small pore size, open stacking pores and highly adjustable porosity [7, 59]. Furthermore, they display superior service life and great dust-holding capacity in various air filtration applications [60].
The use of nano-fibres in the field of air filtration is not recent, as attempts have been made in this direction for over twenty years. In order to achieve an effective filtration, it is necessary to exist an appropriate correspondence between the channels and the structural elements of the filter and the particle sizes or small droplets to be retained by the filter. This is why electrospun nanofibres are very suitable for the creation of filters, because of their very small diameters. Under these conditions, it becomes possible to remove unwanted particles smaller than 1 micron in diameter.

Nanofibres have very high surface area per unit mass, offering an outstanding capacity for the attachment of molecules, ions and different nanometer scale particles: when diameters range from 5 to 500 nm, the surface area per unit mass is varies between 10, 000 to 1,000,000 m²/kg, as it can be seen from figure 6 [61]. Due to the very large specific surface and the high surface cohesion, it is possible to retain particles with dimensions below 0.5 µm, the efficiency of the filtration under these conditions being very high.

Figure 6. Variation of surface area with fibre diameter [61].

One of the decisive characteristics for the performance of the filters is their filtration efficiency, which is closely related to the fineness of the fibres, because, in general, the filtration efficiency increases linearly as the filter membrane compactness decreases and the pressure applied increases. It has been suggested that the size of the fibres in the electrospun filtration layers should be between 50 nm and 500 nm, in order to maintain both filtering efficiency and filter resistance [62].

Another reason for nanofibres filters higher air filtration performance is the difference in the flow of air across the material. Because of the slip flow at the nanofibre surface, the drag force is smaller than for the nonslip flow, causing lower pressure drop. As a result, the fraction of the air flowing near to the surface of fibres is larger, and consequently more particles will transit near the fibre, causing more efficient inertial impaction, interception, and diffusion [63].

6. Polymers used to obtain nanofibres used in air filtration
The durability of the nanofilters in the operational environment depends on their thermal, mechanical, and chemical properties, that depend on the polymer from which the filter is produced [64]. In the last years, numerous polymeric substrates were electrospun to produce nanofibres for air filtration applications. Its is considered that in general the filtration efficiency is improved when the polarity of the polymer is bigger, as the dipole–dipole or dipole–induced dipole force can help the binding of PMs
to the surface of nanofibres. This is why polymers such as polyacrylonitrile, polyvinil alcohol, poly(vinylpyrrolidone), polystyrene are commonly used to make nanofilters [37].

Polyacrylonitrile, a versatile polymer with good physical properties, reasonable price and simple to electrospin [65], was used to produce electrospun nanofibres with diameters in the range of 270–400 nm that have been used as filters that showed high efficiency [50, 66]. Other research showed that electrospun PAN nanofibrous membranes exhibited high air-dust filtration efficiency of more than 99.99% in between PM0.3 and PM2.5 [67]. Electrospun PAN fibres with diameters in 270–400 nm range were used to filter NaCl nanoparticles with diameters below 80 nm and they proved to need less filter mass and showed filtration efficiency compared to commercial filters [68]. Porous bead-on-string filters produced by optimizing the polyacrylonitrile concentration of electrospun dopes and ambient humidity condition in the electrospinning process achieve an efficiency above 99% with a low pressure drop of 27 Pa at an airflow rate of 4.2 cm/s [69]. A thin coat of electrospun PAN nanofibres over a metal mesh showed over 95 % filtration efficiency (PM2.5 in a polluted city environment) with 90 % transparency [70]. TiO2/PAN, ZnO/PAN and Ag/PAN electrospun nanofibres were used as filter media and they maintained high filtration efficiency even after the addition of nanoparticles, and the Ag/PAN nanofibre media had bactericidal activity [71, 72].

Polyvinil alcohol (PVA), a water-soluble synthetic polymer which has outstanding mechanical properties and chemical resistance, was used to produce air filters [73]. Crosslinked electrospun PVA nanofibre membranes increase with about 30% the filtration efficiency of melt-blown substrate layers and realize high removal efficiency for large particles [74]. Nonwoven polypropylene material covered by electrospun PVA nanofibre proved to be capable to eliminate organic polar compounds, ethers and compounds containing carbonyl groups from gas [75].

High-impact polystyrene (HIPS) waste was used to produce nanofibre membranes using electrospinning method and then applied as air filtration media, and the membranes with weight of 12.22 gm-2 displayed an efficiency greater than 99.999%, equivalent to that of the HEPA filter [76].

Nanofibre membranes made by electrospinning waste of acrylonitrile butadiene styrene (ABS), a polymer for widely industrial applications due to its good mechanical and physical properties [77], were used in air filter applications, and the results indicate that ABS waste are good as air filter media for daily needs, but the mechanical property of the membranes can be enhanced placing the membrane between two strong nonwoven membrane [78].

Another most electrospun polymer is polyamide, both 6 and 6.6. Nano-fibres obtained from polyamide 6 via electrospinning were frequently used as filtration media [79], and a very significant parameter in what concerns the filtration efficiency proved to be the solution concentration [80]. The best conditions of electrospinning process were obtained when using a 12.5% polymer solution. The pressure drop increased by increasing the electrospinning time and the thickness of the filter, and the maximum filtration efficiency was 96% for PA nanofibrous filter obtained after 240 min of electrospinning time [11].

Other studies showed that electrospun PA-6 nanofibres have good mechanical properties, and prove good filtration ability for microparticles with up to 0.3 μm diameter [81] and the PA 6 nanofilters had less pressure drop performance compared to HEPA filter [82]. PA 6 filters with 100 nm fibre diameter removed 99.9 % of aerosol particles with 1 μm - 5 μm in diameter from the air flow of 1 m/min [83]. Air filter membranes made from fibres obtained by electrospinning a 21% PA6 solution in formic acid proved to have a filtration efficiency ranging between 93-95% [84]. When using electrospun PA 6 nanofibres of 80–200 nm in diameter, the filtration efficiency was 99.993%, superior to the commercialized HEPA filter at the velocity of 5 cm/s using 0.3 μm test particles [85, 86]. Similar results were obtained for polyamide 6.6, which showed a wider polymer solution electrospinnable range and a wider distribution of the fibre diameter (60–511 nm) compared to polyamide 6 (90–236 nm). In this case a filtration efficiency of 84.9–90.9% was obtained when an 8% PA6.6 % (w/vol) solution has been used [87]. PA-66 filter consisting in traditional nonwoven polypropylene 2D PA-66 spider-web-like nanonets supplied high filtration efficiency (up to 99.9%)
and low pressure drop [88]. Polyamide-56 nanofibre membrane with bimodal structure obtained by electrospinning proved to have good air filtration efficiency [89].

Introducing functional materials into nanofibres may lead to electrospun nanofibres with antibacterial and detoxifying properties. Polyamide 6 electrospun nanofibres were electrospayed with ZnO nanoparticles, and the ZnO/Polyamide nanofibre mats had outstanding antibacterial efficiency against both the Gram-negative Escherichia coli and Gram-positive Bacillus cereus bacteria [90].

Electrospun polyimide nanofibre are applicable for high-temperature air and fume applications, as they have exceptional heat resistance and good flame retardant properties. These filters developed a high-efficiency air filtering, as they captured particulate matter that have a diameter of less than 2.5 micrometers at up to 370°C temperature with over 120 h working time [47].

Another polymer used to manufacture air filters is polylactic acid, which has the additional advantage that it is biodegradable and is derived from renewable resources. Polylactic acid/TiO$_2$ electrospun fibrous membranes showed a filtration efficiency of 99.996% with a relatively low pressure drop (128.7 Pa) and good antibacterial activity [91].

Electrospun polyacrylate air filter membranes captured large particle size (12.07 μm) with efficiencies over 99%, while the smallest (0.095 μm) particles resulted in the lowest capture efficiency of 92.97% [92]. Polyethylene oxide (PEO) was used to obtain nanofibre filters by coating electrospun PEO nanofibres onto a nonwoven microfibre substrate, showing that the most penetrating particle size decreased from 140 to 90 nm and the removal efficiency decreased as the face velocity increased from 5 to 10 cm/s [93].

Fibrous membranes made of electrospun polyvinylidene fluoride (PVDF) with fibre diameter from nanoscale to microscale were used as air filters, and nanofibrous membranes from 10% solution concentration proved to have best air filtration properties [94]. Electrospun cellulose acetate fibres with 0.1–1 μm have been used as filters which demonstrated a slightly better quality factor compared to commercial high efficiency glass fibre at a much lower thickness [95].

Air filters made of electrospun lignin fibres showed unfavorable mechanical properties, but lignin-based composite filters developed with polyethylene oxide (PEO) fibre filters exhibited good filtration efficiency and appropriate mechanical properties [96]. Silk fibroin solution has been electrospun to produce fibroin nanofibre membranes that exhibited a high filtration efficiency of 99.99% with a rather low air resistance of only 75 Pa [97].

Frequently two or more polymers with complementary properties are combined to produce composite polymeric nanofibre membranes which benefit from the merged properties of these polymers [9, 38, 65, 96, 108-119]. These dual/multi component nanofibre filters exhibit many characteristics such as excellent PM2.5 removal efficiency, durability and multi-functionality within a single filter [120-125].

7. Conclusions and further research directions

Nanofibres made by electrospinning are frequently applied in the area of air filtration, as they possess significant advantages such as small and controllable diameter, high porous structure and surface area to volume ratio with internal connectivity, and good mechanical properties. To improve the performance of nanofilters for air filtration, it is necessary to have in-depth knowledge of the mechanisms underlying the separation processes that takes place, namely the relation of the entire set of characteristics of the filter with the specificity of the impurities to be removed. It is essential to know exactly the relevant data that describes the relationship between the structure of the nanofiltration membrane and its transport properties. Although important development has been made in the past decades in the field of electrospinning, further improvements have to be done in what regards the nanofibre properties, such as the specific surface area, the interconnectedness of the fibres, and their thermal and mechanical properties, and this is why more fundamental experimental and theoretical analysis of the process is needed. In the future, the research will also focus on the production of functional filters, capable of selectively capture targets of interest, such as VOCs, PM2.5, viruses and many other contaminants.
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