

FINE PARTICLE REMOVAL USING HYDROPHOBIC MICROPOROUS POLYMERIC MEMBRANES

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Article history

Received

8 March 2018

Received in revised form

5 November 2018

Accepted

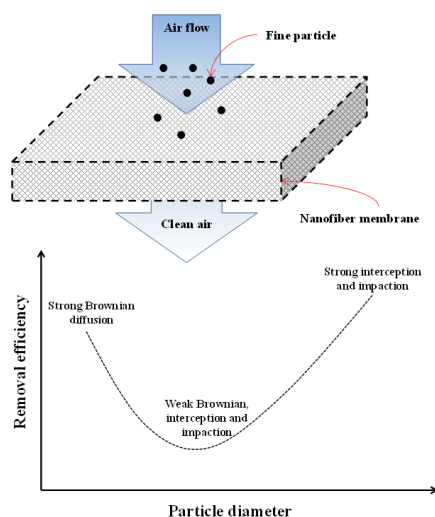
10 March 2019

Published online

18 April 2019

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Graphical abstract



Abstract

The air quality in the world has been worsening in the last decades due to industrial, vehicle, cigarettes smoke, forest fire, and fuel usage. In this case, fine particles are the world's greatest concern due to its aerodynamic properties which enable it to travel throughout the world. The current conventional technologies seem to have lost their reliability due to complexity, low removal efficiency, and high equipment cost. Membrane air filter brings new hope to answer this challenge. It gives high removal efficiency with an acceptable pressure drop to fulfill the need for clean air at a lower price. Recently, the introduction of nanofibre membrane as a low-cost membrane may broaden membrane application in air filtration. Compared to conventional membrane, nanofibre membrane offers some interesting features such as higher porosity, interconnected pore structure, and narrow pore size distribution that provide remarkable permeability. In this paper, the microporous polymeric membrane for air filtration especially for fine particles removal is reviewed including mechanism of fine particle removal, membrane preparation, and factor affecting filtration performance.

Keywords: Air filter, fine particles, hydrophobic, nanofiber, polymeric membrane

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1.0 INTRODUCTION

The air quality in the world has been worsening from year-to-year due to air pollution. There are several factors that drive this phenomenon such as industrial, vehicle, cigarettes smoke, forest fire, and fuel usage [1]. Basically, air pollution happens due to the changes in chemical, physical, and biological properties of air that brings detrimental effects upon living creatures [2]. Air pollution can stimulate respiratory disease, eye irritation, throat irritation, or even bronchitis, and asthma. In addition, it can also induce the death of plants because of acid rain [3]. Therefore, controlling particulate matters in the air is essential to overcome this problem [4].

In air pollution, fine particles are the greatest health concern due to their ability to pass through

the nose and throat, and be absorbed deep inside the lungs, thus causing more harmful effects [5]. Exposure to fine particles can cause short-term health effects such as eye, nose, throat and lung irritation, coughing, sneezing, runny nose, and shortness of breath. Fine particles are defined as airborne particles with a diameter of 2.5 μm or less (PM_{2.5}). Considering its aerodynamic properties, fine particles can be carried by wind from one place to another easily.

Several technologies have been developed to minimise removal of fine particles, such as electrostatic precipitation, cold plasma, wet scrubbing, cyclonic air filtration (venturi), and physical filters (glass fibres, melt-blown fibres, spun-bonded fibres, and ceramic filters) [6-8]. However, most of these technologies are not capable to

provide energy-efficient as well as cost-effective filtration. Membrane air filter brings new hope to answer this challenge. Membrane filtration allows clean air to pass, yet rejects fine particles present in polluted air [9-14].

Polymeric materials have long been developed for membrane gas separation due to its lightweight, easy preparation procedure, and low cost. Polyvinylidene fluoride [15], polysulfone [16], polylactic acid [17, 18], polyacrylonitrile [19-21], polyethersulfone [22], polyetherimide [18], and cellulose acetate [23] are examples of materials that have been used to prepare membrane for fine particles removal. These membranes have excellent filtration efficiency, more than 99%. For better understanding, this paper aims to give an overview of the microporous polymeric membrane technologies for fine particles removal. Membrane preparation, properties, performance, and challenges are also discussed.

2.0 MECHANISMS OF FINE PARTICLES REMOVAL IN MEMBRANE BASED AIR FILTRATION

Nowadays, membranes have been used for several applications including respiratory protection, air sampling, clean rooms, industrial processes, and building ventilation system. Generally, a membrane for air filtration consists of dense material or mat which captures and retains particles throughout its

depth or thickness. There are a lot of membranes with different materials that have been manufactured with features such as small pore volume and hydrophilic or hydrophobic properties. Moreover, many modifications can also be made to enhance the removal efficiency as well as lower the pressure drop across the membrane.

There is a common understanding that the separation process by membrane occurred by sieving mechanism. In sieving mechanism, particles which are larger than the membrane pore are trapped, while the smaller ones pass through. However, this mechanism is only applied for liquids; it is absolutely not true in the air filtration [24]. In this section, the mechanism of particulate filtration in the air is described to avoid this misconception.

Several types of physical mechanisms can be used for high-efficiency particulate removal. The most influential mechanisms are straining, interception, inertial impaction, and Brownian diffusion [25]. The first mechanism, straining, is actually similar to sieving mechanism in liquid. It occurs when the smallest size of particles is larger than the membrane pore size. The mechanism of straining is shown in Figure 1(b). Meanwhile, Figure 1(c) shows the interception mechanism. Interception happens when a particle in a gas stream comes within one particle radius of a filter fibre [24]. For a certain number of particle size, there are streamlines which move near the membrane fibre so that the particles are intercepted. If the particles are too far from the fibre, they do not contribute to this mechanism.

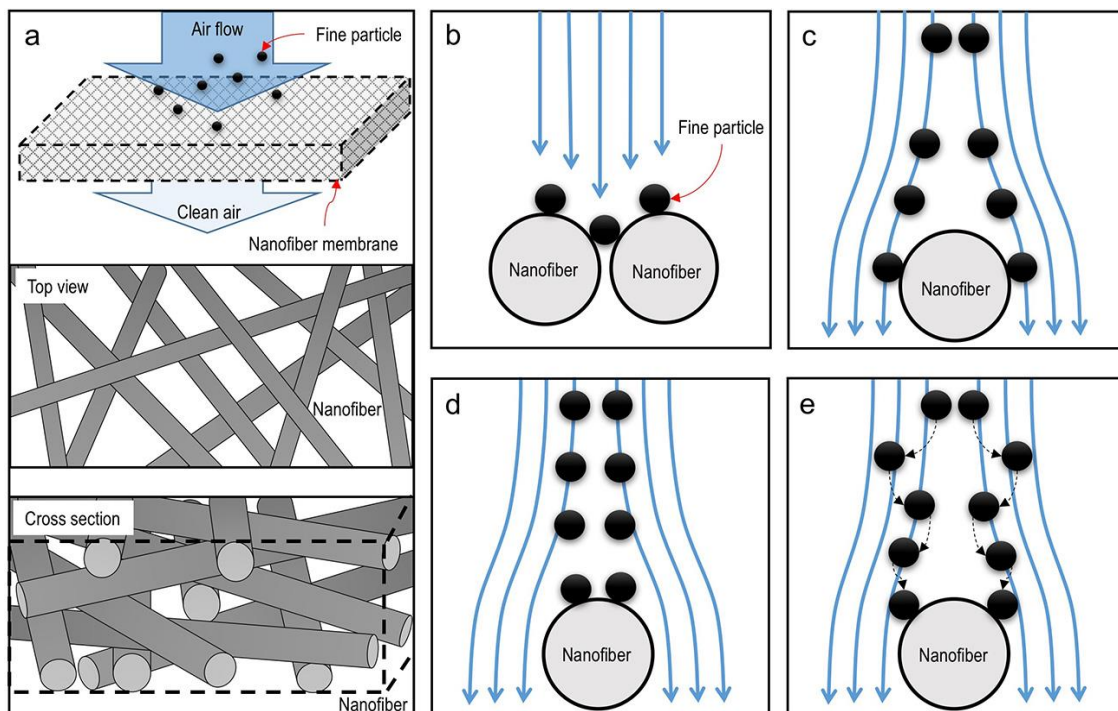


Figure 1 Illustration of fine particles removal in nanofiber membrane: (a) nanofiber membrane, top view, and cross section, (b) straining/sieving; (c) interception; (d) impaction; (e) Brownian diffusion

The next mechanism is called inertial impaction. It occurs when a particle is too large, thus it is unable to quickly follow the changes in streamlines direction near the filter. This mechanism is dominant while the velocity is high, and the filter has high-density packing. The illustration of impaction is shown in Figure 1(d). Furthermore, the last mechanism happens in low gas velocities as well as small particles with diameters of 0.1 μm or below. Those particles tend to make a random motion. This random motion is widely known as Brownian diffusion. The illustration of particulate removal capture in this mechanism is shown in Figure 1(e).

3.0 DEVELOPMENT OF MEMBRANE BASED AIR FILTRATION

The selection of membrane materials is one of the most important tasks in membrane separation technology. Membranes for air filtration with different materials as well as the modifications have been studied by several researchers. The major findings for each type of materials and modifications are summarized in Table 1.

Table 1 Materials and modifications of membrane for fine particulates removal

No	Materials	Modification	Pore size	Particulate size	Filtration Efficiency	Pressure Drop	Ref.
1	Polyvinylidene fluoride	Al ₂ O ₃	0.36 μm	-	99.17%	-	[15]
2	Polyvinylidene fluoride	-	0.56 μm	-	94.20%	-	[15]
3	Polysulfone	TiO ₂	-	0.3-0.5 μm	99.9%	40 Pa	[16]
4	Polylactic acid	Nano-sized/porous comp.	2-120 nm	0.26 μm	99.9999%	95 Pa	[17]
5	Polylactic acid	-	2.9-3.6 μm	0.26 μm	99.997%	165 Pa	[18]
6	Polyimide	Carbon Woven	3.3 μm	0.3 - 2.5 μm	93%	462.56 Pa	[26]
7	Polyethersulfone/ Polyamide 66	-	-	0.3-1 μm	99.999%	537 Pa	[22]
8	Polyethersulfone	-	-	0.3-1 μm	92.45%	513 Pa	[22]
9	Polyetherimide	Silica Fibrous SiO ₂	3.46 μm	0.3 μm	99.99%	50 Pa	[18]
10	Polyetherimide	-	4.11 μm	0.3 μm	99.85%	58 Pa	[18]
11	Polyetherimide	Nanoparticles Boehmite	-	0.3 μm	95%	75 Pa	[18]
12	Polyetherimide	Nanoparticles Si ₃ N ₄	-	0.3 μm	82%	60 Pa	[18]
13	Polyetherimide	Nanoparticles BaTiO ₃	-	0.3 μm	85%	58 Pa	[18]
14	Polyamide-6/ Polyacrylonitrile	Nanofibre/nets	0.3-0.4 μm	0.3-0.5 μm	99.999%	105 Pa	[27]
15	Polyamide-56	Nanofibre/nets	0.5-0.7 μm	0.3-0.5 μm	99.996%	100 Pa	[28]
16	Polyamide-56	-	-	0.3-1 μm	97.76%	482 Pa	[22]
17	Polyacrylonitrile	Silica Nanofibrous SiO ₂	0.01-0.06 μm	0.3-0.5 μm	99.99%	117 Pa	[19]
18	Polyacrylonitrile	-	-	0.05-0.08 μm	99.99%	220 Pa	[20]
19	Polyacrylonitrile/ Polyurethane	FPU	1.49 μm	0.3-0.5 μm	99.98%	110 Pa	[21]
20	Polyvinylacetate	-	-	0.3-0.5 μm	97.38%	132.8 Pa	[23]
21	Polymethylmethacrylate	-	0.39 μm	0.02-0.3 μm	99.8%	13 Pa	[29]
22	Soy protein / bacterial cellulose	Acrylic acid	-	0.1-10 μm	99.94%	-	[30]
23	Polyamide6/ Polytrimethylene T	-	0.1 μm	0.1-0.4 μm	95%	59 Pa	[31]
24	Polyvinylalcohol	Silica nanofibrous	25-50 nm	0.3-0.5 μm	99.7%	100 Pa	[32]
25	Cellulose acetate	-	-	0.3-0.5 μm	88.59%	139.9 Pa	[23]
26	Carbon woven	High voltage	-	-	89.60%	36.8 Pa	[33]

It can be seen that a lot of hydrophobic polymers have been used as potential materials to produce air filtration membranes. These materials also offer advantages of their stability. For instance, polysulfone is known for their toughness and stability at high temperatures. Meanwhile, polyimide has excellent physicochemical properties such as high temperature stability, exceptional mechanical strength, dimensional stability, superior insulation properties, and radiation and chemical resistance. Polyvinylidene fluoride (PVDF) is stable while it is attacked by most of the corrosive chemicals and organic compounds including acids, alkaline, strong oxidants, and halogens [34]. In addition, polyetherimide and polyacrylonitrile have been widely approved as membrane materials with good chemical and thermal stability, and excellent separation properties.

Several modifications have also been extensively studied to improve membrane performance that indicated by removal efficiency and pressure drop. The modifications consisted of nanoparticles addition, combination with nanofibre/nets, composite membrane, high voltage usage, and modification of membrane structure (e.g., sandwich, multilayer, etc.).

3.1 Polymeric Membrane for Fine Particles Removal

For air separation, asymmetric membranes are usually used. These membranes consist of a dense skin layer with a thickness of 0.1–0.5 μm and supported by a porous sub-layer with a thickness of about 50–150 μm [35]. Both the skin and sub-layer are composed of the same material. Skin layer has a role as the actual separating barrier, while the sub-

layer acts only as a mechanical support. However, for several polymers such as polypropylene is commonly produced as symmetric membrane.

The most common method for fabricating asymmetric membrane is phase inversion. In this method, the membrane morphology is determined by the phase separation, and subsequent solidification of the cast polymer solution. Membrane preparation using phase inversion method can be conducted by dry process, wet process, or a combination of dry-wet process [36]. The schematic diagram of dry, wet, and dry-wet process is shown in Figure 2. The properties of the resultant membrane are mainly controlled by polymer concentration, solvent ratio, evaporation time, and rheological factors [37], which then greatly affect its separation performance.

To obtain high permeability and selectivity, several modifications have been investigated, such as polymer blending and crosslinking modification. Polymer blending is a simple method to modify polymer properties. In this method, a certain polymer solution is generally mixed with another polymer in different proportions and the resultant solution is used for the preparation of the membrane [38, 39]. An important aspect that needs to be considered is miscibility, i.e., the ability of the polymers to form a homogenous mixture. Meanwhile, cross-linking modification has been widely used to improve the membrane selectivity. The enhanced selectivity is due to the restriction of mobility, and an increase in packing density of polymer chains. Cross-linking can be induced by UV irradiation, ion beam irradiation, plasma treatment, thermal treatment, and chemical compounds [40–44].

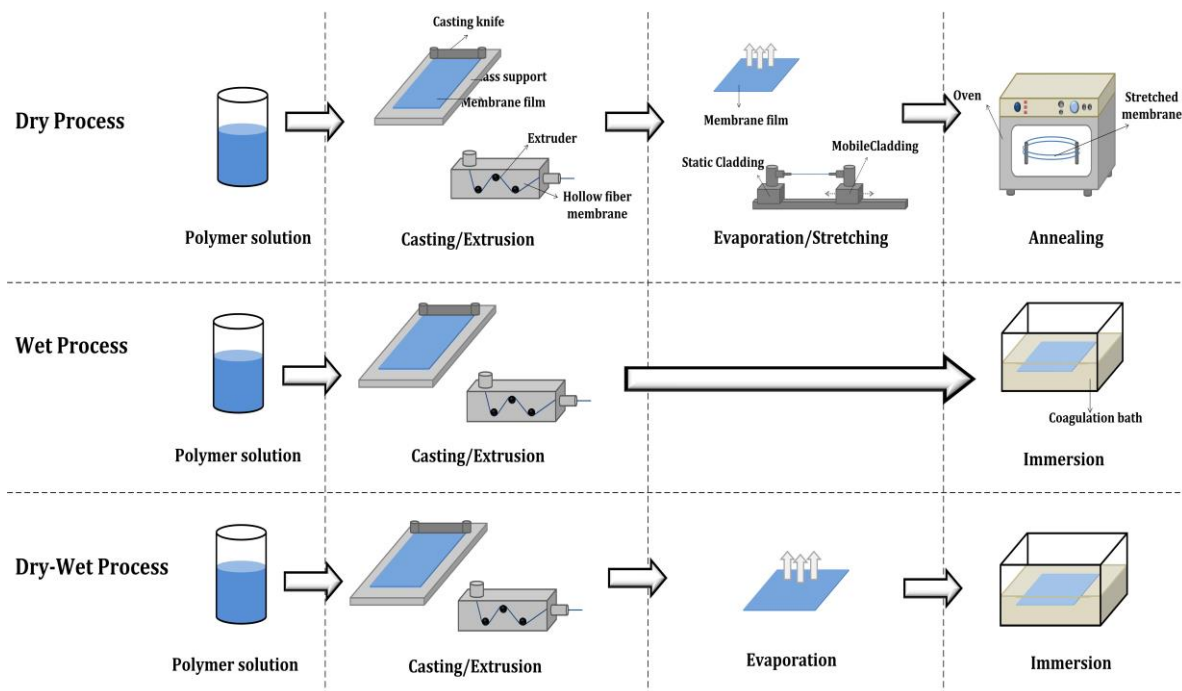


Figure 2 Schematic diagram of dry, wet, and dry-wet process

3.2 Nanofiber Membrane for Fine Particles Removal

Fine particles with a diameter of 2.5 μm or less have high deposition in the alveolar region during mouth breathing [45]. Besides, bio-aerosols with sizes 0.3 to 30 μm also became a big problem if they entered human respiratory system [46]. Several air separation technologies still have not been able to remove the small particles. One of the promising technologies for this case is nanofiber membrane. Nanofibers have several advantages, such as high density of pores, high surface area to volume ratio, high permeability, flexibility in surface functionalities, and superior mechanical performance [47, 48]. These outstanding properties make nanofiber membranes suitable to be used as filter media in air filtration applications.

Nanofiber membrane is defined as hollow fibre membrane with a diameter of less than 0.1 μm . Several techniques have been developed to produce nanofibres, such as melt fibrillation, gas jet techniques, nanolithography, and self-assembly [49–51]. However, these techniques are limited by combinations of restricted material ranges, possible

fibre assembly, cost, and production rate. In recent years, electrospinning had been chosen to replace previous techniques. Electrospinning uses high voltages to produce an interconnected membrane-like web of small fibres (10–500 nm in diameter) from electrostatically charged polymer solutions [38, 52, 53]. The fibres were produced from the plastic stretching of a jet of polymeric solution, while the solvent evaporated, and the polymer solidified at micro and nanoscales on a grounded surface or collector [54]. The schematic illustration of electrospinning process is shown in Figure 3.

Electrospinning is applicable to a wide range of polymers like those used in conventional spinning, i.e., polyolefin, polyamides, polyester, aramid, acrylic as well as biopolymers like proteins, DNA, polypeptides, or others like electric conducting and photonic polymers [55]. Gopal *et al.* [56] successfully produced polyvinylidene difluoride (PVDF) nanofiber membranes for microfiltration of polystyrene particles. The results showed a high rejection rate of more than 90% of polystyrene micro-particles.

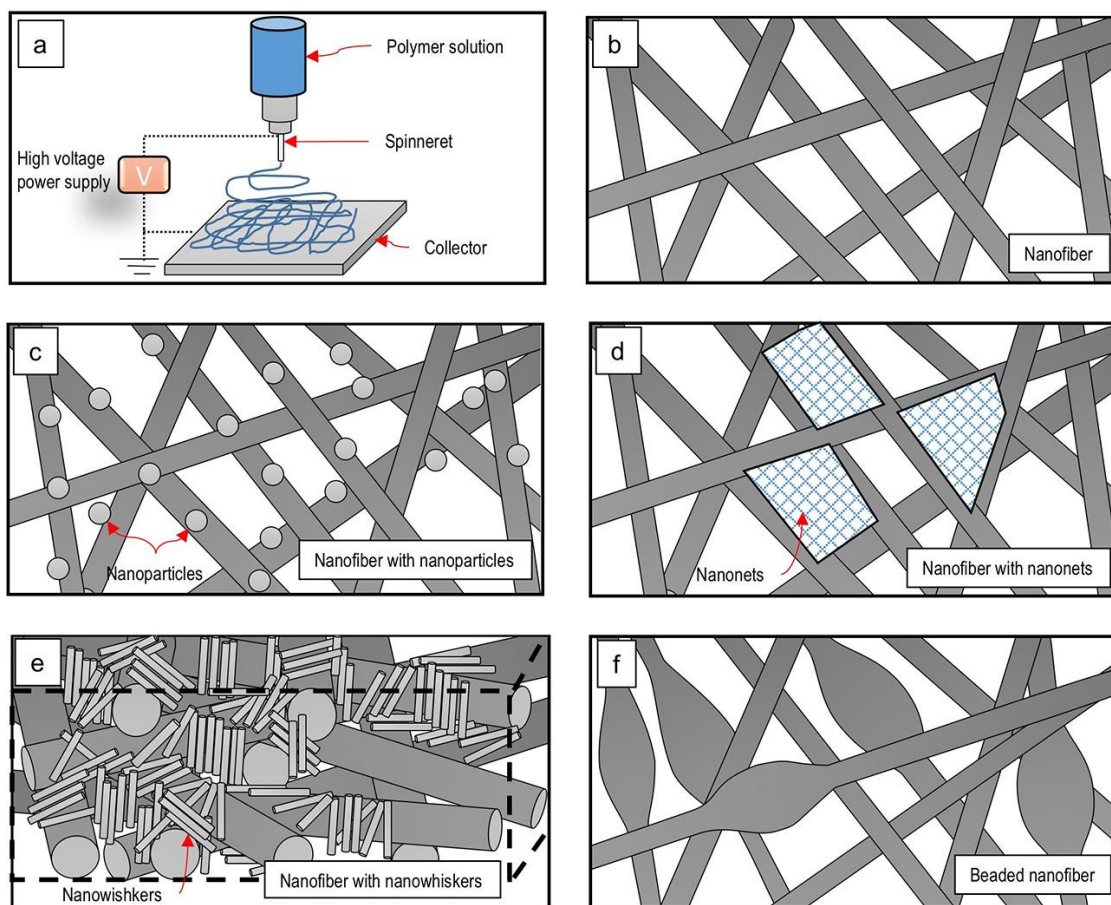


Figure 3 Schematic illustration of (a) electrospinning process, (b) nanofiber membrane (top view), (c) nanofiber with nanoparticles, (d) nanofiber with nanonets, (e) nanofiber with nanowiskers, and (f) beaded nanofiber

In general, nanofibre membranes still have some drawbacks, which include inadequate filtration performance, weak mechanical property, and short life caused by the deep bed filtration manner [28]. To optimise the performance of nanofibre membrane, the incorporation of nanoparticles such as MgO, TiO₂, Al₂O₃, and other oxides was recently explored by Sundarrajan *et al.* [57-59]. These nanoparticles were selected due to their unique ability to decontaminate wide varieties of toxic gases, such as chemical contaminants, biological contaminants (viruses, bacteria), pesticides, and many more. After mixing the nanoparticles with polymer solutions followed by electrospinning, the obtained nanocomposite membranes were tested for decontamination of the stimulant of nerve gas, paraoxon. From the pressure drop analysis, it was found that an effective decrease in pressure drop (up to 34%) occurred throughout the multilayer membrane as a function of the raised nanoparticle content [58].

Furthermore, several studies had combined nanofibre with nanonets or nanowhiskers. Liu *et al.* [28] successfully produced polyamide-56 nanofibre/nets membrane with bimodal structures. The addition of nanonets increased the mechanical strength of the membrane as well as the filtration efficiency up to 99.9995%. Meanwhile, Zhang *et al.* [46] used micro-wave structured polyamide-6 nanofibre/nets membrane with embedded poly(m-phenylene isophthalamide) (PMIA) staple fibres for effective ultrafine particle filtration. The results showed that the addition of nanonets led to increased filtration efficiency (99%), and a decrease of pressure drop (60 Pa).

In addition, improvement of nanofibre for air filtration could also be conducted by producing beaded nanofibre. Yang *et al.* [27] used a sandwich structured polyamide-6/polyacrylonitrile nanonets/bead-on string composite membrane. The resulted membrane had high separation efficiency up to 99.9998% with low pressure drop, 117.5 Pa for 300-500 nm airborne particles. Wang *et al.* [18] also obtained high filtration efficiency (99.997%) using porous bead-on-string poly-(lactic acid) (PLA) fibrous membranes. The increase of PLA led to the decrease in beads number, while the beads size increased. With increased beads size, volume fraction of fibrous membrane decreased. Thus, there was an increase in membrane permeability, and a decrease in pressure drop.

Nanofibre structure formed by electrospinning strongly depends on the solution parameters, such as polymer molecular weight, polymer concentration, the blend ratios, viscosity, conductivity, and choice of the solvent [48, 54, 60]. Fong *et al.* [61] found that viscosities 1-20 poises and surface tension 35- 55

dynes/cm were suitable for poly(ethylene oxide) (PEO) fibre formation. Electrospinning was prohibited at viscosities above 20 poises due to the instability of flow caused by the high cohesiveness of the solution. Meanwhile, Liu and Hsieh [62] found that viscosities 1.2-10.2 poises were applicable for cellulose acetate (CA).

Furthermore, one of the most important quantities related to electrospinning is the fibre diameter. A higher viscosity led to a larger fibre diameter [60, 61, 63]. The fibre diameter also increased with increasing polymer concentration [64, 65]. In addition, the applied electrical voltage also has a remarkable effect on the fibre diameter. A higher applied voltage ejected more fluid in a jet, resulting in a larger fibre diameter [64].

4.0 FACTORS AFFECTING MEMBRANE PERFORMANCE

Besides the internal factors (materials and modifications), there were several external factors that should be considered while operating membranes for fine particles removal. The first common parameter is air flow rate. Basically, the air flow does not affect the removal efficiency if the pore size is smaller than the particle size considering the straining mechanism [28].

When the pore size was larger than the particle size, the increased air flow reduced filtration efficiency. As the flow rate increased, retention time of the particles moving through the membrane became lower, and this increased the possibility of the deposited particles to be removed due to the higher shear force. These phenomena reduced the chance of particles deposited by Brownian diffusion, interception, or impaction [26]. It was also observed that no matter what mechanism occurred, the increase in flow rate always increased the pressure drop of the membrane. The effect of flow rate on the pressure drop for different membrane materials is shown in Figure 4. Other factors that also contributed to the membrane performance were temperature and pressure. If the temperature or pressure was too high i.e., beyond its thermal stability or tensile strength, the membrane distorted causing the pore size to become larger. At this phase, the removal efficiency decreased significantly [66].

The particle diameter also affected membrane performance. However, it only happened when the separation followed interception, impaction, and Brownian diffusion mechanism. On the other hand, if straining was the only mechanism that occurred during the filtration, the particle diameter had no effect towards the removal efficiency.

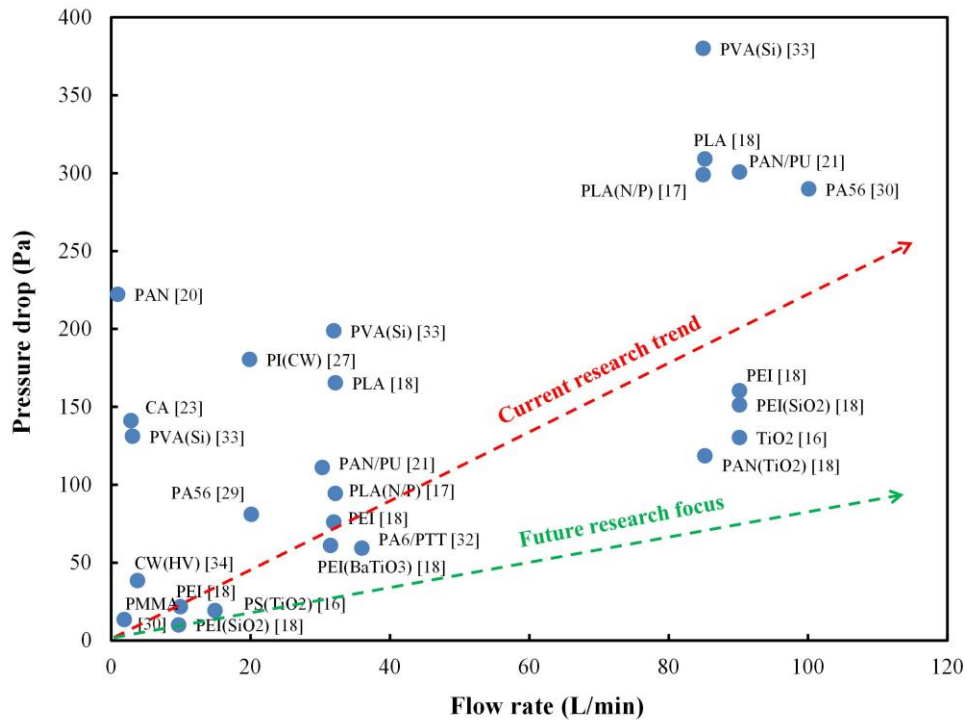


Figure 4 Effect of air flow rate to the pressure drop

Furthermore, particle diameter significantly affected removal efficiency. At first, as the particle diameter increased, the removal efficiency decreased. However, after it had reached the lowest point also known as the most abundant penetration particle size (MPPS) point, the removal efficiency increased with the increase of particle diameter. For the lowest particle size, the Brownian diffusion was the most dominant mechanism. When the diameter became larger, the mechanism started to change into interception and impaction, and finally at MPPS point, the effects of those mechanisms were at their weakest point, allowing the particle to go through the membrane easily. Nevertheless, after the MPPS point, the removal efficiency improved due to the stronger effect of inception and direct impaction mechanism [67]. In brief, after discussing the factors that affected membrane performance, it is necessary to optimise all sensitive parameters to fabricate an ideal membrane for fine particles removal.

5.0 CONCLUSION

The need for clean air is increasing day by day. A large variety of technologies to produce clean air

from the polluted one have been evaluated. However, conventional technologies seem to have reached its saturation point of improvement. In contrast, due to its younger age, membrane based air filtration showed superior performance. It can be operated in a wide range of operating conditions such as flow rate, particle diameter, and concentration.

Furthermore, the use of nanofibre membranes can be an effective technology to remove small particles. It is important to avoid unwanted respiratory disorders by hazardous small size particles. In addition, several operational factors have been proven to influence the performance of membrane for air filtration, such as air flow rate, temperature, pressure, and particle diameters. In brief, after discussing the factors that affect membrane performance, it is necessary to optimise all sensitive parameters to fabricate an ideal membrane for fine particles removal. In addition, the weak mechanical strength of nanofibre membrane should be taken into consideration for future development.

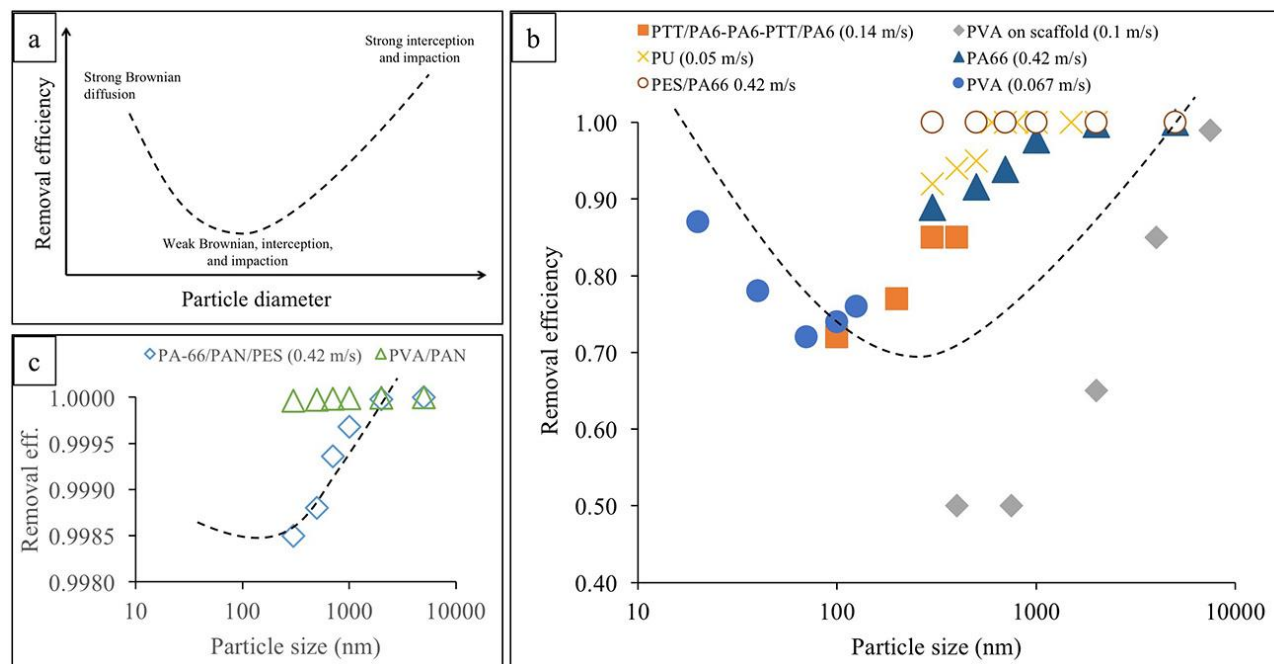


Figure 5 The effect of particle diameter on filtration efficiency. (a) Trends, (b) and (c) typical example of nanofibre membrane removal efficiencies [31, 68-73]

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