Fabrication and characterization of electrospun PAN-Ag nanofiber membranes for antibacterial activity and air nanofiltration

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***Abstract*— Particulate matter and airborne microorganisms are two of the most serious indoor air problems due to their significant risks to human health. Comprehensive research on air filtration with good filtration performance for fine particles and antibacterial function is essential. In this study, after trial and error and optimization of conditions, polyacrylonitrile (PAN) 10% - 1% silver nanoparticles (AgNPs) membranes with suitable morphology and uniform diameter distribution successfully designed and fabricated by an electrospinning method. These electrospun mats showed good antibacterial activity toward Staphylococcus aureus (Gram-positive bacteria) and Escherichia coli (Gram-negative bacteria). With integrated properties of small fiber diameter and robust mechanical strength of 7.14 MPa, the resultant PAN10%-1%Ag membranes exhibits high filtration efficiency of 99.27%, low pressure drop of 33 Pa, and good quality factor. The fabrication of PAN-Ag membranes will have broad applications, including face mask, indoor air filtration and clean room.**

Keywords*— *Nanofibers; Electrospinning; air Filtration; antibacterial activity, membrane

I. Introduction

People expend a lot of time in buildings, and indoor air quality (IAQ) is tightly related to people's health, work and even lives. Particulate matter (PM) and airborne microorganism has attracted more attention following the increasing trend of severe environmental problems, due to the increased emissions of automobile exhaust and industrial air, as well as frequent sandstorm accidents [1]. Technically, PM2.5 refers to particulate matter 2.5 µm or less in diameter. PM2.5 and submicron particles are noticeably harmful and can migrate to the human lungs or in some cases to other organs of the body, thus increasing respiratory and cardiac morbidity [2,3]. PM2.5 combined of organic matter (organic carbon and elemental carbon) and inorganic components (such as SiO2, SO4, and NO3) [4,5]. Additionally, the airborne microorganism that is intercepted on the surface of the fibrous air filters do not die but can multiply under suitable conditions of temperature and relative humidity, and if the filter is broken, the microorganisms will enter the indoor environment [6]. As we all know, electrospinning is known as a simple and effective technique to fabricate polymer nanofibers [7]. Polymer nanofibers have attracted the attention of many researchers in recent decades due to their high specific surface area, small pore size and special features that are interesting in advanced applications [8-10].

The objective of this study is to evaluate and develop a composite filter made of polyacrylonitrile (PAN) nanofibers with incorporated silver (Ag) nanoparticles on polyethylene terephthalate (PET) nonwoven fabric mesh. Also, the morphology, mechanical properties, filtration performance and antibacterial activity of nanofiber membranes have been carefully investigated.

II. Material and Methods

PAN powders (Mw = 90,000) were purchased from Spectrum Chemicals and Laboratory Products Co., Ltd., USA. N, N-dimethylformamide (DMF) was obtained from Shanghai Chemical Reagents Co., Ltd, China. Silver nitrate (MW 169.88 g/mol) was purchased from SRL chemicals, India. The polyethylene terephthalate (PET) nonwoven fabric mesh with negligible filtration capacity (filtration efficiency of ~3.5% and pressure drop of ~0.5 Pa for 300 nm particles under the face velocity of 32 L/min) for fiber receiving was purchased from the commercial market. All chemicals were used without further purification.

10 wt% (note that wt1% is equal to 1g of PAN powders in 100g of DMF) solutions were prepared by dissolving PAN in DMF with a magnetic stirring process for 24 h at room temperature. The fabrication of nanofibrous membranes was performed by using the DXES-3 spinning equipment (Shanghai Oriental Flying Nanotechnology Co., Ltd., China). Typically, homogeneous solutions are loaded into 5 ml plastic syringes and injected through 24-gauge metal needles at a feed rate for electrospinning. The stainless-steel roller covered with a non-woven polyethylene terephthalate substrate rotates at a speed of 80 rpm and keeping a tip-to-collector distance of 20 cm. During the electrospinning process, flow rates of PAN were controlled at 0.2 ml/h, by a peristaltic pump. The high voltage applied to the needle of the PAN solutions syringe was 18 kV, the relevant temperature and humidity were 25 ± 2 0C and 33 ± 2%, respectively. All samples were vacuum-dried at 70 0C for 1 h to remove the residual solvent and charges. schematic of the experimental set-up is shown in **Fig.1**.

The morphology of the composite membranes was examined by field emission scanning electron microscopy (FESEM; S-4800, Hitachi Ltd, Japan). The fiber diameters in the membrane were measured at 50 different points by the image analyzer (digimizer software). Antibacterial activity of

PAN-AgNPs electrospun membranes was investigated by the zone inhibition method. Staphylococcus aureus (Gram-positive bacteria) and Escherichia coli (Gram-negative bacteria) were chosen as model microorganisms in this study. The mechanical properties of the different samples were measured with a testing machine (Instron 3345, single column, UK) at a crosshead speed of 20 mm/min at room temperature, the membranes tested had a length of 30 mm and a width of 5 mm. Atmospheric aerosol was used as experimental particles. A pump (Model DING HWA Co) assures the air circulation in the device. The upstream and downstream aerosol concentration was determined by a condensation nucleus particle counter (Model 5.412, GRIMM Co). The membrane pressure drop was obtained by a pressure manometer device (Model 202, KIMO Co).

III. Results and Discussion

Optimizing the membrane design to achieve the highest filtration performance of fine particles requires the creation of membranes with small fiber diameter and low packing density. The SEM images for PAN nanofibers membranes with 0,0.5,1 and 2 wt.% Ag nanoparticles are shown in **Fig.2 (a)** to **(d)** which shows the distribution of nanofiber diameters. From the **Fig.2**, it is determined that the nanofibers are randomly arranged with uniform diameter. There are no droplet or bead on the surface of the fibers, indicating that the electrospinning parameters have been selected logically in this study. In nanofibrous membranes, a three-dimensional porous structure is observed due to the large number of pores between fibers.

The antibacterial property of PAN-AgNPs with different contents of AgNPs (0, 0.5, 1 and 2 wt %) against Escherichia coli and Staphylococcus aureus as shown in **Fig.3** and **Fig.4** that performed by the disc diffusion susceptibility test after 24 h incubation. The diameter of inhibition zone for each sample was recorded in the **Table 1**. As observed in zone of inhibition data, Pristine PAN the diameter of inhibition zone for pure PAN is 6.0 mm and no change was observed in the pure PAN membranes antibacterial efficiency (the diameter of the cut discs of the membranes for inhibition zone test is 6 mm). The inhibition zones of PAN-AgNPs (0.5, 1.0 and 2.0 wt %) against Escherichia coli were 7.05 ± 0.3, 8.45 ± 0.9 and 7.35 ± 0.5 mm, respectively.

Furthermore, PAN-AgNPs (0.5 and 1.0 wt %) were exhibited to inhibit the growth of bacteria with slightly higher effectiveness against Staphylococcus aureus compared with Escherichia coli, the zones of inhibition increased to 7.61± 0.1 and 9.51± 0.4 mm, which indicating Gram-positive bacteria might be more sensitive to silver antibacterial agents than Gram-negative bacteria. It was mentioned that the most antibacterial effected against for both two microorganisms when PAN-1%Ag loaded to PAN electrospun membranes. After adding more AgNPs, the antibacterial property of PAN electrospun membranes weakened mainly owing to the potentially devastating effects of AgNPs and to the aggregation of AgNPs at relatively high concentrations [11].

The mechanical properties of membranes in severe operating conditions such as high air flow and working pressure have some weaknesses in the filtration applications. The PAN10%-1%Ag electrospun membranes fabricated at 10 wt % exhibited a relatively high tensile strength of 7.14 MPa, an elongation at break value of 31.73%, and a tensile modulus of 119.45 MPa.

The mechanical properties result of PAN10%-1%Ag shows the resulting membrane has a good mechanical property compared to similar nanofibers and has the ability to applied in air filtration applications [12].

Considering the proportionality between filtration efficiency and pressure drop, it seems that PAN10%-1%Ag nanofibers with a basis weight of 1.50 g/m^2 are the most suitable options for selecting optimal nanofibers, which are used in other air filtration tests.

As can be seen in **Fig. 5a**, it was observed that as the particle size increases, the filtration efficiency first decreases to reach the minimum filtration efficiency and then increases. Nevertheless, it is interesting to note that PAN10%-1%Ag is more effective in trapping the particles. The filtration efficiency is 99.27%,84.81% and 95.27% for the 100 ,300 and 500 nm aerosol particles. Therefore, PAN10%-1%Ag membrane has a greater ability to remove aerosol particles.

Fig. 5b shows the pressure drop versus face velocity curves, the slope of which can be used to assess the air permeability of filters.

Fig. 5c that could be determined by the following equation to exhaustively assess the filtration performance of the PAN10%-1%Ag membrane was presented.

$$QF = -\ln(1-\eta)/\Delta P \qquad (1)$$

where η and ΔP represented the filtration efficiency and pressure drop, respectively [13,14]. Filter with a better filtration performance should have a higher efficiency and a higher QF.

It is observed that QF of PAN10%-1%Ag membrane is better in contrast to other filtration media, which could be attributed to small fiber diameter, small pore size and highly porosity of resulted nanofiber structure shows the more effective application of this filter against airborne pollutants and SARS-CoV-2 and a great application potential in the energy saving society.

IV. CONCLUSION

In summary, after trial and error and optimization of conditions, PAN10%-1%Ag membrane with with suitable morphology and uniform diameter distribution successfully designed and fabricated by electrospinning. These electrospun mats showed good mechanical property and antibacterial activity toward Staphylococcus aureus (Gram-positive bacteria) and Escherichia coli (Gram-negative bacteria). The resulting membrane exhibit robust mechanical strength, high filtration efficiency of 99.27% for the 100 nm aerosol particles, less pressure drop of 33 Pa and good quality factor for airborne particles. The basis weight of a PAN-Ag nanofiber membrane is only 1.50 g/m^2, 1/54th of a typical commercial mask with similar filtration efficiency. It is anticipated that the PAN-Ag membranes will have broad applications, including face mask, indoor air filtration and clean room.

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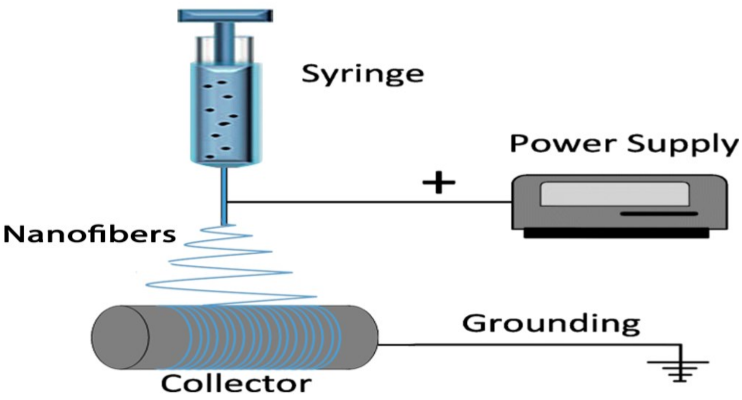

Fig 1. Schematic showing the fabrication of PAN-AgNPs electrospun nanofibers on the collector drum.

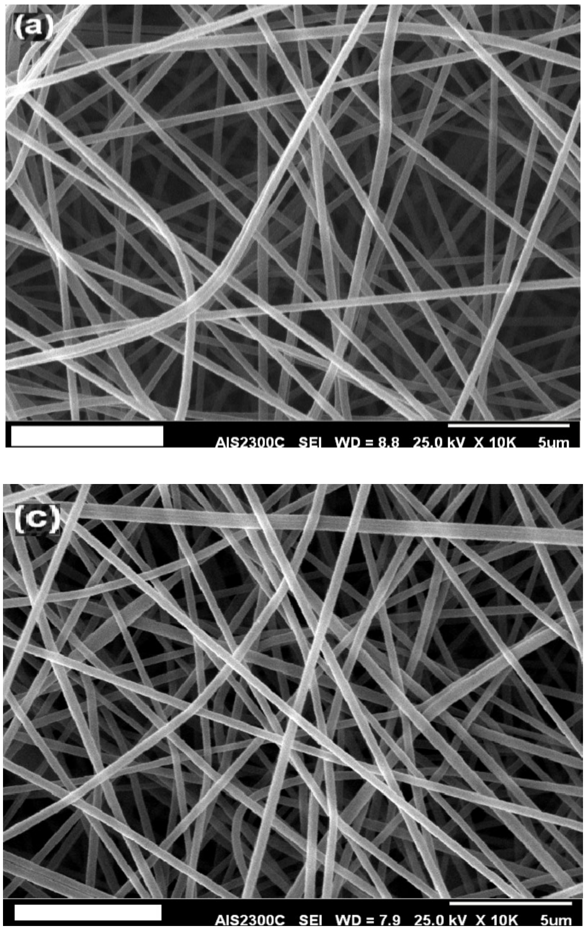

Fig 2. SEM images and fiber diameter distribution of nanofibers; **(a)** PAN 10%, 0% Ag; **(b)** PAN 10%, 0.5% Ag ; **(c)** PAN 10%,1% Ag; **(d)** PAN 10%, 2% Ag.

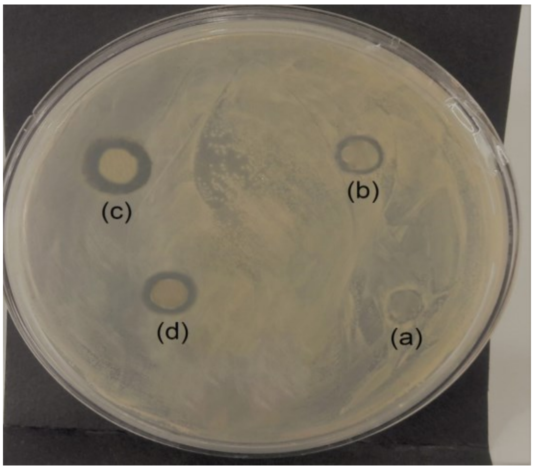

Fig 3. Photograph showing zone of inhibition of the PAN-Ag membranes against Escherichia coli included **a)** Ag 0.0%, **b)** Ag 0.5%, **c)** Ag 1.0% and **d)** Ag 2.0%.

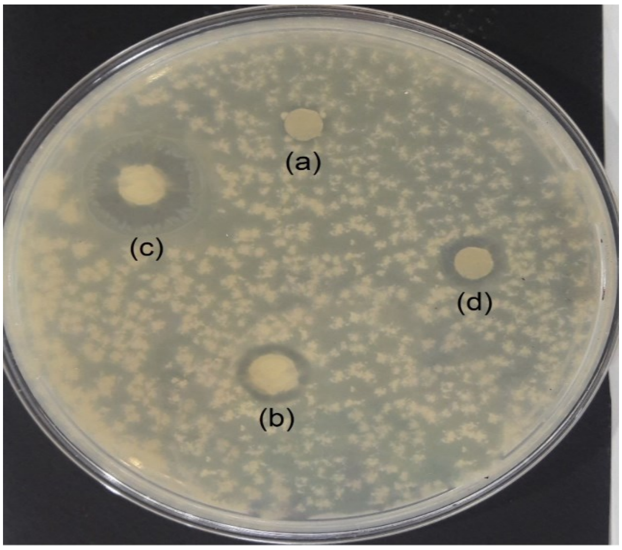

Fig 4. Photograph showing zone of inhibition of the PAN-Ag membranes against staphylococcus aureus included **a)** Ag 0.0%, **b)** Ag 0.5%, **c)** Ag 1.0% and **d)** Ag 2.0%.

Table 1. The diameter of Inhibition zones of PAN-Ag electrospun nanofibers against Escherichia coli and Staphylococcus aureus.

	Escherichia coli (Gram$^-$)	Staphylococcus aureus (Gram$^+$)
Sample	**inhibition zone (mm)**	**Inhibition Zone (mm)**
Ag 0.0	6.0 ± 0.0	6.0 ± 0.0
Ag 0.5	7.05 ± 0.3	7.61 ± 0.1
Ag 1.0	8.45 ± 0.9	9.51 ± 0.4
Ag 2.0	7.35 ± 0.5	7.74 ± 0.2

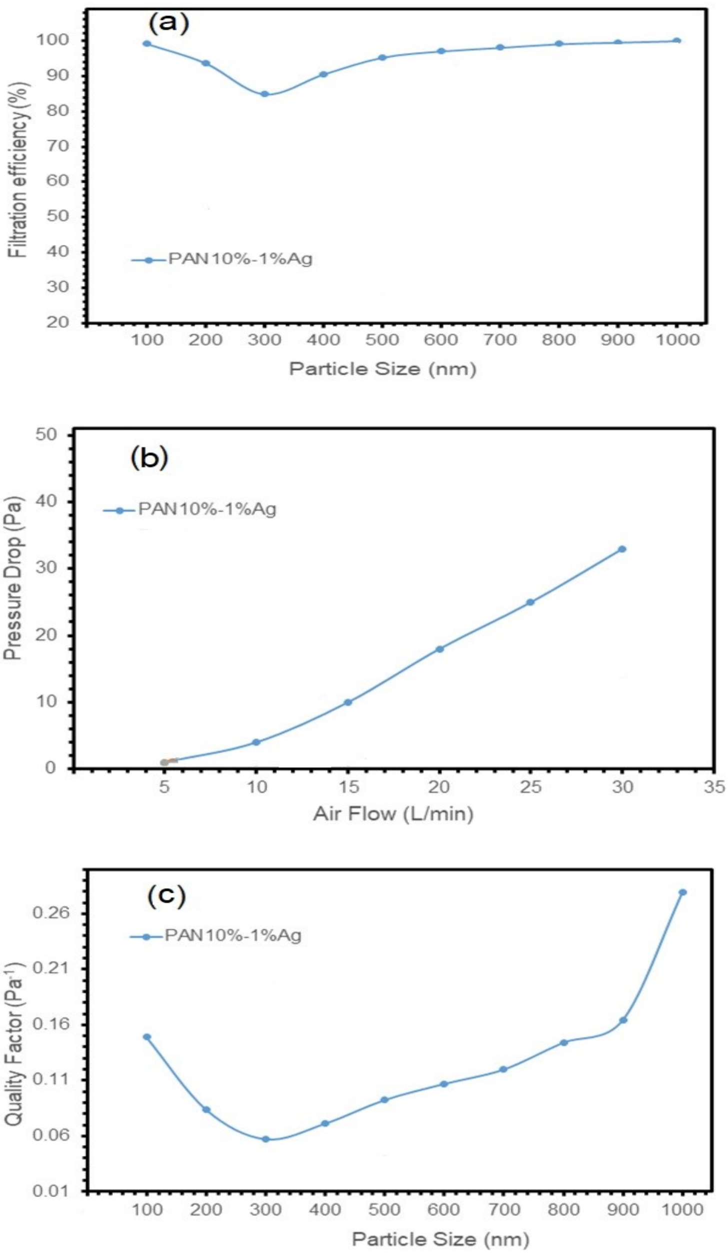

Fig 5. (a) Filtration efficiency of PAN10%-1%Ag (1.50 g/m^2), **(b)** pressure drop versus face velocity of the PAN10% -1%Ag, **(c)** quality factor of PAN10% -1%Ag.