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Pore engineering towards highly efficient electrospun nanofibrous membranes for aerosol particle removal

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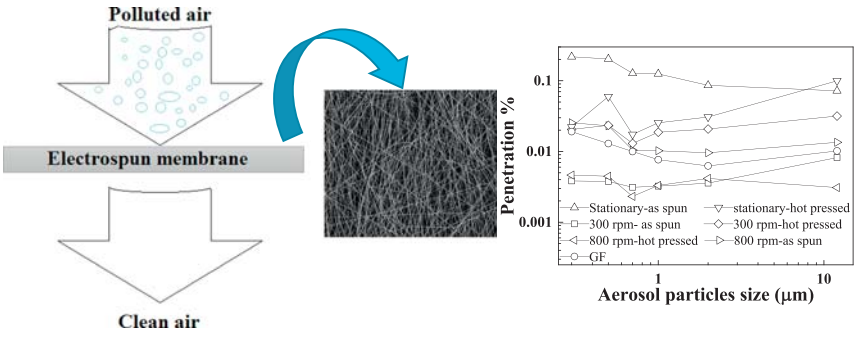
HIGHLIGHTS

- Electrospun PAN based nanofiber membranes are emerging materials for air purification.
- The controlled inter-fibre spacing and orientation design is the key to optimize filtration performance.
- The samples were benchmarked against commercial filters and found to exhibit higher air filtration efficiency.
- Processing techniques correlated differences in pore morphologies, fibre orientation, and filtration performance.

GRAPHICAL ABSTRACT



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ABSTRACT

Electrospun nanofibrous membranes were engineered for aerosol particle removal by controlling the fiber density and alignment across electrospun mats. Electrospun nanofiber membranes were deposited on both, rotatory drum and stationary collectors, to investigate the effect of fiber alignment on filtration performance. Poly(acrylonitrile)/dimethyl formamide (PAN/DMF) solutions were used to produce membranes for applications in air purification. The air filtration performance of as-produced and hot-compacted membranes were systematically evaluated with regard to penetration, pressure drop, and quality factor when subjected to potassium chloride (KCl) aerosol particles in the size-range of 300 nm to 12 μm. The membranes offered air filtration efficiencies in the range of 77.7% to 99.616% and quality factors between 0.0026 and 0.0204 (1/Pa). The samples were benchmarked against commercial filters and were found to exhibit similar quality factors but higher air filtration efficiencies. These results were correlated to differences in pore morphologies and fiber orientation distributions generated from the different processing techniques, which revealed that the alteration of the fiber density is an effective method for enhancing air filtration performance.

1. Introduction

Concerns arising from increasing exposure to high concentrations of air pollutants, including particulate matter (PM), powders or fumes produced from industrial activities, vehicles exhaust systems or energy

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production plants, are triggering discussions and initiatives to implement stricter governmental regulations for the protection of human health (Homaeigohar and Elbahri, 2014; Samet and Gruskin, 2015; Montefusco, 2005; Chuanfang, 2012). Air pollutants have been demonstrated to be lead causes for lung cancer, respiratory infections, allergies, asthma, heart failure, and cardiovascular diseases, whose impact is exacerbated and particularly prevalent in dense urban developments (Mannucci et al., 2015; Mentz and O'Brien, 2016; Miller et al., 2007).

Membrane filtration is nowadays considered to be the most efficient and reliable physical method for protection from air pollutants (Givehchi and Tan, 2015). The filtration market is estimated to reach US$700 billion by 2020 (Huang et al., 2003). Traditional commercial air filters such as melt-blown based fibrous filters and glass fiber filters offer low separation efficiencies for particles in the range of 100 to 500 nm due to the relatively large, micrometre scale, of the fibers compared to the airborne particles (Wang et al., 2016). In order to enhance the air filtration performance of non-woven type membranes, it is in these cases necessary to increase the thickness of membranes, which dramatically increases the pressure drop of the membranes (Zhu et al., 2016).

Nanofiber membranes formed by electrospinning offer elegant and extremely efficient solutions to the above-mentioned drawbacks of conventional commercial filters. The electrospinning technology allows to produce very fine fiber diameters (Fan et al., 2016), in the range of 40 and 2000 nm, from a large array of polymers (Park and Park, 2005; Kosmider and Scott, 2002), while the resulting highly interconnected fiber mat morphologies offer very high specific surface area to volume ratios (Sridhar et al., 2015), as well as tuneable porosity and pore size distributions (Lu and Ding, 2008; Bhardwaj and Kundu, 2010; Wang et al., 2013). Highly efficient air filtration membranes have been developed from various biopolymers and synthetics polymers, such as, polyamide 6 (PA6) (Vitchuli et al., 2010), polyacrylonitrile (PAN) (Liu et al., 2015), polyvinyl alcohol (PVA) (Wang et al., 2010), polyurethane (PU) (Scholten et al., 2011), keratin (Aluigi et al., 2009), or chitosan (Desai et al., 2009). Furthermore, electrospun air filtration membranes may be further enhanced through specific decoration or functionalization with additives or modifiers such as silicon dioxide (SiO_2) (Wang et al., 2014), aluminium oxide (Al_2O_3), and titanium dioxide (TiO_2) (Chuang et al., 2014) to enhance the air pollutants filtration capacity while also increasing the mats mechanical, thermal, and chemical resistance.

In the present work, a novel and facile approach is developed to control the structure of air filtration membranes and improve their air filtration performance in terms of filtration efficiency, pressure drop and quality factor (QF). The manipulation of fiber alignment and density by using different fiber-collection methods, including rotatory and static collectors, has allowed engineering diverse pore morphologies from a different fiber size distribution. In addition, the impact of fiber density was also investigated by hot-pressing selected as-spun membranes to alter the packing density of the material and the fiber-to-fiber spacing. The effect of fiber alignment on the resulting porosity, pore size distribution, overall fiber packing density, alignment and interconnectivity of the membranes allowed to engineer mechanically stronger and more efficient membrane materials.

The key to the novel strategy is to benefit from the controlled inter-fiber spacing and orientation design to optimize interactions of the particles with the fiber surfaces, shifting the predominant capture mechanism from sieving to particle interception. This effect is shown to occur due to the interception of fine particles occurring primarily when the sub-micron particles get in contact with the surface of fibrous membrane materials and adhere to them due to Van der Waals interactions. The new strategy provides a promising solution to generate ultra-thin membranes for capturing fine, sub-300 nm, particles from air, and is an up-scalable option towards the production of cheap and efficient large-scale separation materials.

2. Experimental part

2.1. Materials

Poly(acrylonitrile) (PAN) (Mw = 150,000 g/mol) powder and N-N dimethylformamide (DMF) of 99.8% purity were used for the electrospinning solutions to synthesize the nanofibers, and potassium chloride (KCl) of 99.9% purity for the generation of aerosol particles. All chemicals were purchased from Sigma-Aldrich and used without further treatment.

2.2. Fabrication of electrospun nanofiber membranes

Solutions of 8 and 10 wt% of PAN were dissolved in DMF to prepare the precursor electrospinning solutions. The solutions were stirred for 12 h at room temperature to produce a homogeneous dope. A lab-designed electrospinning system previously described (Bagherzadeh et al., 2014) with a stationary collector, and a commercial electrospinning unit from HOLMARK HO-NFES-043U (Kalamassery, Kerala, India) with a rotational drum collector (operating in the range between 300–900 rotations per minutes (rpm)) were used to produce the electrospun membranes. The choice of polymer concentrations was based on previous work (Al-Attabi et al., 2017) and the solutions were placed in a plastic syringe fitted with a metal needle (Gauge 23) used as an electrospinning nozzle. Three different conditions (stationary collector, drum with 300 rpm, and drum with 800 rpm) were used to collect the fibers spun from the nozzle.

The as-spun electrospun membranes were then divided into two series. The first series of electrospun membranes were hot-pressed at 95 °C for 2 h with a 1.7 kPa on hot press model (YH-380). The second series of electrospun membranes were characterized and used as-spun without further treatment. The electrospinning operating conditions are shown in Table 1. The basis weight for the electrospun fiber mats electrospun for a fixed 2 h duration using a single syringe system were calculated based on the amount of polymer discharged and the area that was covered by the web. These values are displayed in Table 1.

2.3. Scanning electron microscopy

Scanning Electron Micrographs (SEMs) were acquired on a JEOL 7800F (JEOL, Tokyo, Japan). The SEMs of electrospun samples were used to examine the morphology of the electrospun membranes and to evaluate both, pore and fiber distributions, as well as to determine overall compaction levels from hot pressing. The accelerating voltage was typically of 5 kV for a working distance of 10 mm. A BAL-TEC SCD 050 sputter coater (Leica Microsystems, Australia) was used to coat the samples with a 5-nm gold layer to prevent charging during imaging. The image analysis software “Image J” was used to determine the fiber diameter and alignment of the electrospun membranes from at least 3 SEMs per sample. Fast Fourier transform (FFT) was used for the purpose of fiber alignment evaluation. The technique in details was described by (Both Engel et al., 2015). The original SEMs was converted to FFT images using “Image J” analysis software to show the gray scale pixels, which were distributed in different patterns according to the fiber alignment. Then the frequency of the pixel intensity of FFT images was plotted as a function of radial angle ranging between 0 and 36° using the Oval Profile plugin.

2.4. Pore size distribution

The pore size distributions of the membranes were evaluated using a capillary flow porometer (3gzh Quantachrome, Florida, Untied States of America). The process was described in a previous work (Dumée et al., 2011). The mean pore size, pore size distribution, and bubble point pressure were measured over a pressure range of 0.25–0.75 bar and with Porofil as a wetting liquid.