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Fabrication of low-cost and high-performance coal fly ash nanofibrous membranes via electrospinning for the control of harmful substances

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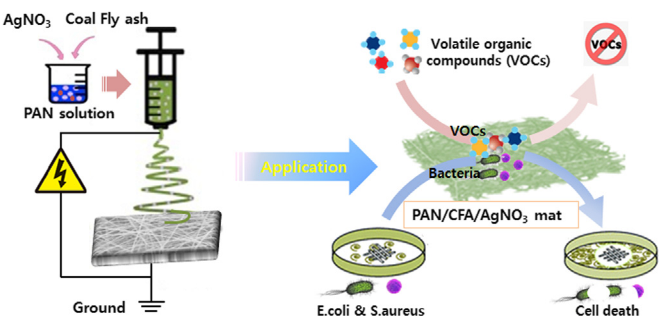
GRAPHICAL ABSTRACT



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ABSTRACT

The use of a high-performance and low-cost electrospun nanofiber membrane to remove harmful pollutants from the air is highly valuable in terms of commercial applications. In this paper three functional materials were successfully fabricated into a low-cost, high-performance fibrous mat via a simple and one-step electrospinning technology. These materials included coal fly ash (CFA), which is an industrial waste mainly derived from coal-fired power plants with an adsorption capacity for adsorbing of volatile organic compounds (VOCs); silver nitrate ($AgNO_3$), which imparts excellent antibacterial ability; and polyacrylonitrile (PAN), which serves as an electrospun substrate with high spin-ability. The binding properties of different amounts of CFA powder, $AgNO_3$ and PAN were observed and analyzed in detail by field emission scanning electron microscopy (FE-SEM), biological transmission electron microscopy (Bio-TEM), X-ray diffraction (XRD) and other experimental analyses. The results show that the PAN nanofibers containing 40 wt% CFA (relative to the weight of PAN) had the highest VOCs adsorption capacity compared with other membranes. Furthermore, 2 wt% $AgNO_3$ (relative to the weight of PAN) and 40 wt% CFA powder can be easily fabricated with and PAN nanofibers, and this electrospun PAN/CFA/$AgNO_3$ composite membrane has excellent antibacterial ability on Gram-negative *Escherichia coli* (*E. coli*) (ATCC 52922) and Gram-positive *Staphylococcus aureus* (*S. aureus*) (ATCC 29231) bacteria. These electrospun fibrous mats currently stand as an emerging material that has huge development potential for treatment of environmental pollutants.

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1. Introduction

Since Reneker's group found electrospinning could be adopted to produce various polymeric nanofibers by using a wide range of polymer solutions under the roles of electric field force and surface tension, electrospinning technology has become more and more focused [1,2]. Especially in recent decades, electrospinning has become one of the main ways for the effective preparation of nanofiber materials due to its manufacture simplicity, low cost spinning, spinning a wide variety of materials, and process controllability [3]. With the development of nanotechnology, electrospinning, as a simple and effective novel processing technology capable of producing nanofibers with smaller pores and higher surface area, plays an important role in various fields, such as, filtration application, environmental engineering, biomedical application (e.g. medical prostheses, pharmaceutical, tissue template, wound dressing), protective clothing application, electrical and optical application and other functional application [4,5]. Among them, filtration application for removing harmful substances in air or water with various electrospun nanofibers has received widespread attention from many researchers in recent years, due to its advantages of high porosity and filtration efficiency, porous structure, good tensile strength, low-cost, less synthetic steps, good repeatability, green synthesis process and others [6–8].

Volatile organic compounds (VOCs) are organic chemicals with a low boiling point and high vapor pressure. They comprise one of the major air pollutants, mainly arising from petroleum refineries, the combustion of fossil fuels, chemical industries, automobile industries, paint industries, and building materials [9]. In addition to anthropogenic sources, there are some natural sources of VOCs produced from biogenic emissions of terrestrial and oceanic environments [10]. Some VOCs, such as chloroform, benzene, toluene and xylene are harmful to human health and the ecological environment. Common yet highly toxic and carcinogenic VOCs include aldehydes, aromatic compounds and polycyclic aromatic hydrocarbons (PAHs) [11,12]. They can cause various respiratory problems and increase the chance of cell cancerization [13]. Some VOCs have photochemical reactions with NOx and sulfur dioxide (SO_2), forming ozone and other photochemical smog in the troposphere, resulting in both stratospheric ozone layer depletion and Antarctic Ozone Hole (AOH) formation [14,15].

Therefore, the control and reduction of VOCs in the air is necessary. Reducing the use of fossil fuels in vehicles, such as through the development of electric vehicles (EV) or those that use other alternative fuels, is one of the most effective ways to reduce VOCs in this sector [16,17]. Many researchers have pointed out that the amount of VOCs emerging from diesel engines fueled with biodiesel fuel or biodiesel blended with diesel fuel is much less than that resulting with the combustion of pure diesel fuel [18,19]. In addition to reducing VOCs by either using lower amounts of traditional fuel and switching to alternative fuels, other approaches have been used, including incineration, catalytic oxidation, biodegradation, absorption, adsorption, membrane separation and other post-processing control methods [10]. Among them, the adsorption method is the most economical and effective means to reduce and control the emission of VOCs into the air [20,21].

Activated carbon (AC) is one of the most common adsorbents and is often used to adsorb harmful substances in the air, such as benzene, aldehyde ketones, alcohols, and hydrocarbons. Activated carbon has the advantages of a large specific surface area, moderate pore size, good porosity, uniform pore distribution, fast adsorption speed, and good chemical stability [10,22]. However, AC poorly adsorbs larger VOCs (pores size > nm) because most of their pore structures are microporous, about 2 nm [10]. Moreover, AC has several disadvantages, including pores and channels that are easily blocked by dust; high cost; fire risk; hygroscopicity; early malfunction with some component mixtures; unsuitability for use with wet flue gases; limited application of powdered AC; risk of polymerization by unsaturated hydrocarbons on the AC, which is exothermic and causes blockages; and additional problems associated with regeneration [23–25].

For these reasons, cheaper and more efficient alternative materials have been investigated to remove harmful VOCs from the air. One such material is coal fly ash (CFA), which is a product of coal-fired power plants. Coal fly ash itself is an environmental pollutant and will cause serious air and water pollution without proper handling of direct discharge [26]. On the other hand, CFA has many physical properties similar to AC, including a high surface area, porosity and high adsorption capacity. Therefore, it can be modified to serve as an inexpensive and high-performing adsorbent for treatment of environment pollutants [27–29]. Electrospinning technology has received considerable interest for fabricating nanocomposite membranes with high specific surface areas and a wide range of fiber diameters ranging from the nanoscale to micrometer scale [11]. In addition, efficient disposal of CFA is a worldwide issue because of its massive volume and harmful effects on the environment [30]. As a simple and economical technique for recycling CFA, fabrication of multi-functional fibrous membranes from CFA by electrospinning has attracted a great deal of attention. In terms of antibacterial activity, silver nanoparticles have been widely used as an antimicrobial agent for the synthesis of various antibacterial materials [31]. Many researchers have reported that silver nanoparticles have a good antibacterial effect on various species of bacteria, including Escherichia coli, Staphylococcus aureus, Streptococcus mutans and Pseudomonas mendocina [32,33]. Thus, in this study, CFA powder, silver nitrate ($AgNO_3$) and polyacrylonitrile (PAN) were successfully fabricated into a multi-functional membrane via electrospinning technology, and their combined characteristics were investigated. We evaluated the ability of the multi-functional electrospun PAN/CFA/$AgNO_3$ fibrous membranes to adsorb VOCs from the air and investigated antibacterial efficacy toward Gram-negative Escherichia coli (*E. coli*) (ATCC 52922) and Gram-positive Staphylococcus aureus (*S. aureus*) (ATCC 29231) bacteria. The PAN fibers containing 40 wt% CFA powder and 2 wt% $AgNO_3$ NPs (relative to the weight of PAN) had the highest VOCs adsorption capacity and antibacterial ability compared to other electrospun membranes. The use of CFA is environmentally friendly because it takes waste material from industry and effectively uses it to create a multifunctional and high-value material. Electrospun PAN/CFA/$AgNO_3$ fibrous membranes can be used in a variety of filter applications for cleaning air pollutants and have great potential for applications in controlling pollutants.

2. Materials and methods

2.1. Materials

Polyacrylonitrile (PAN) and N,N dimethylformamide (DMF) were used as-received and purchased from Sigma Aldrich Co., USA, and Samchun Chemical Co., Ltd., Korea, respectively. CFA powder, which was used as an adsorbent for adsorption of VOCs, was purchased from Won Engineering Company Ltd., Korea. Toluene and xylene solutions were purchased from Sigma Aldrich Co., USA, and benzene solution was purchased from Samchun Chemical Co., Ltd., Korea. $AgNO_3$ crystal powder was purchased from Sigma Aldrich and was used as an antibacterial agent against *E. coli* and *S. aureus* bacteria.

2.2. Characterization of electrospun fibrous mat

Field emission scanning electron microscopy (FE-SEM, JIB-4601F, JEOL Ltd., Tokyo, Japan) was used to observe the morphology of the as-prepared fibrous mats. Biological transmission electron microscopy (Bio-TEM, Hitachi Tem H-7650, Hitachi, Ltd., Tokyo, Japan) was similarly used to observe the interlayer structure of the as-prepared composite fibrous mats. N_2 adsorption-desorption isotherms were investigated at 77 K by an ASAP 2020 (V2.09) physisorption analyzer (Micromeritics Co., Norcross, GA, USA). The chemical composition of the as-prepared composite fibrous mats were confirmed via Fourier