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Production of Nanofibers Containing Magnesium Oxide Nanoparticles for the Purpose of Bioaerosol Removal

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ABSTRACT: The present study aims at investigation of the performance of nanofibrous filter, containing magnesium oxide (MgO) nanoparticles, for bioaerosols removal from the air stream. It synthesizes two types of polyacrylonitrile (PAN) and PAN/MgO nanofibers via electrospinning technique, and investigates the antibacterial properties of the produced nanofibers through disk diffusion. The air containing staphylococcus epidermidis is introduced into the filter test rig by a nebulizer and air sampling from the microorganisms takes place before and after the filters by means of a cascade impactor with blood agar culture medium, with the filters, themselves, examined at two states of UVC radiation and dark. The mean diameters of PAN/MgO and PAN are 221.38±65.56 nm and 320.25±87.35 nm, respectively, with the mean length of the inhibition zone for these nanofibers calculated as 0 (for PAN) and 2.8 mm (for PAN/MgO). It turns out that the mean percentage of filtration efficiency is higher in case of PAN/MgO than PAN nanofiber filter; however, the former displays higher mean pressure drop than the latter. For both types of nanofibers under UVC radiation, the mean percentage efficiency for bioaerosol removal is higher than in the dark.

Keywords: Nanofibers; Nanoparticle; Air Filter; Bioaerosol; Removal Efficiency.

INTRODUCTION

The bioaerosols at workplaces are hazardous agent, which receive less attention from researchers, in comparison to other occupational hazardous agents. Bioaerosols refer to airborne particles that include living organisms such as bacteria, viruses, and fungi, as well as their metabolites (Kim et al., 2017). They have considerable health effects and the potential health hazards from their exposure along with the progressive need for protection of human workforce and workers against these particles has become evident nowadays. Presence of bacteria and fungi in the air can cause respiratory infections, simultaneously spreading contagious diseases. Exposure to microorganisms needs the particles to find an aerosol form, rendering their distribution a necessary condition (Hakansson et al., 2018). Studies have shown that around 5-34% of air pollution is related to the presence of bioaerosols (Morakinyo et al., 2016).

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Occupational exposure to bioaerosols could occurred in workplaces like agriculture, compost making, and waste management, with high concentrations of airborne bacteria and fungi, resulting in respiratory diseases like allergies and infections (Walser et al., 2015) (Matuka et al., 2018).

There are various technologies to remove these microorganisms from the air stream. Yet, new approaches are still needed to control these biological pollutants at the workplaces. In this regard, filtration is one of the most effective and simplest air purification methods. Currently, it is very common to use High Efficiency Particulate Air (HEPA) filter in air pollution control devices, since their performance is suitable for separating micron particles form air, which can not only improve the surrounding air quality but reduce the complications of diseases, associated with microbial agents (Pigeot-Remy et al., 2014). However, in order to enhance the filtration efficiency especially for submicron to nano particles, the size of fiber should be reduced down to nano scale (Ozden and Basal, 2017). The use of nano-fibrous filters is an economical and efficient method to remove pollutants from the air stream that can bring about greater productivity and higher performance in filtration, compared to microfiber (Somayeh Farhang Dehghan et al., 2016). Currently, nano-fibrous filters have attracted a great deal of attention in air purification processes, thanks to their low cost as well as porous structures, with high permeability and small pore (Mohraz et al., 2018).

The air filters in heating, ventilation, and air conditioning (HVAC) systems mostly operate under dark and humid conditions at room temperature, suitable for the growth of bacteria, mold, and fungi. This gets even worse when these microorganisms attach to the particles, accumulated on the filter, which they even consume as food, thus increasing the accumulation further. Consequently, air quality drops unpredictably, generating a foul odor (Lala et al., 2007) (Burge, 2018). Hence, there have been some attempts to modify the surface of filter media with antimicrobial agents with a long durability, leading to the publication of papers, related to nanofiber filters with antimicrobial properties as well (De Faria et al., 2015). However, microorganisms' accumulation on the filter could reduce their contact with antimicrobial agents, which can be largely solved through the entrance of metal oxide nanoparticles (Tobler and Warner, 2005). Their positive charge ion leads to the absorption of the bacterial cells' electric charge, causing the cell membrane or bacterial DNA to attach to the sulfhydryl group, which prevents the proliferation of microorganisms (Son et al., 2004). This is due to the unique potentials of these nanoparticles to clear a wide range of biological contaminants (viruses, bacteria), pesticides, and many others. The common metal oxide nanocrystals are highly efficient in contamination removal, being considered active absorbents for many chemical and biological agents. The composites of metal oxide particles get into contact with the target substance to absorb it, remove the contamination, or neutralize it. The affinity of metal oxide nanoparticles with important biological components such as sulfhydryl, amino, imidazole, carboxyl, and phosphate groups is the primary cause of their antimicrobial activity (Balamurugan et al., 2011; Ravikumar et al., 2011).

Few studies have assessed the airborne bioaerosol removal by nanofibers (Li et al., 2009; Pham and Lee, 2016), nevertheless, their antibacterial properties have been studied frequently (Kong and Jang, 2008; Shalumon et al., 2011). Accordingly, the present research tried to produce a polymer nanofibrous filter, containing magnesium oxide (MgO) nanoparticles via electrospinning technique and investigate its performance for removal of bioaerosols from the air stream in a filter test rig. Further, the antibacterial properties of the produced nanofibers were also studied. For the first time in the present study, the photocatalytic and antibacterial properties of MgO/PAN hybrid nanofibers has been assessed.

MATERIAL AND METHODS

Nonwoven nanofibrous filters were produced by electrospinning process. To prepare the electrospinning solution, magnesium oxide (MgO) nanoparticles (size: 20 nm, Merck Co., Darmsatdt, Germany) were mixed with polyacrylonitrile (PAN) polymer (molecular weight: 80000 g/mol, Polyacryle Co. (Isfahan, Iran)), aided by N, N-dimethyl formamide (DMF) solution with a ratio of 1:3. This mixture was then stirred at room temperature for 12-24 h to achieve a homogenous solution by a magnetic stirrer and ultrasonic bath. Two samples PAN and PAN/MgO nanofibers were synthesized under electrospinning conditions, which include solution concentration = 16 wt%, applied voltage = 20 kV, nozzle-to-collector distance = 10 cm, air temperature = 20-25^{0}C, electrospinning time = 180 min, injection rate = 1 mL/h, needle diameter = 1.2 mm, and rotating drum speed = 500-1000 rpm (SF Dehghan et al., 2016; Dehghan et al., 2015).

The antibacterial properties of the produced nanofibers were investigated through disk diffusion or Kirbty Bauer method, according to which, latex bacteria of staphylococcus aureus were prepared with a turbidity standard of 0.5 McFarland (1.5*10^{8} CFU/ml). It was then scattered by a swab across the plate surface (150 mm), containing Muller-Hinton agar growth medium, 5 mm thick. Thereafter, blank discs, which contained a certain concentration (10 µlit of the initial concentration 10%) of the tested materials, were placed on the plate surface. After being placed in an incubator (Innova-4000; USA) at 37^{0}C for one night, the diameter of the circle, formed in response to lack of bacteria growth around the disc was measured. Once the plates from the incubator were withdrawn, the width of the inhibition zone of the bacteria was measured to each disc piece, using a digital micrometer (International Organization for Standardization (ISO), 2004).

Fig. 1 demonstrates the schematic design of the filter test rig for measuring the bioaerosol removal efficiency by nanofibrous filter. The tunnel of interest was around 2 m long overall. Made of galvanized sheet, it had a plenum chamber, 80 cm long as well as a channel, 20 cm wide. The bacterial strains were prepared as frozen and got cultured in blood agar medium. Next, they were kept in an incubator at 37^{0}C for 24 h. Then, the required amounts of bacterial colony were taken from the medium by a loop and got dissolved in oral NaCl serum 9%. The concentration of the resulting suspension, which ought to reach 0.5 McFarland, was examined with the absorption extent or passage percentage, as read by spectrophotometer (Unico Spectro Quest Model SQ2800 Single Beam UV / Visible Scanning Spectrophotometer; Canada).

To do the test, the air containing staphylococcus epidermidis (with a concentration of 10% and 0.5 McFarland as well as a 98.7% passage equivalent to 10^{7} CFU/ml) was introduced into this tunnel by a nebulizer (KUN 808 King Ultrasonic; Taiwan), having a pressure of 7-12 psi. The sampling from the microorganisms was done in a test tunnel by a cascade impactor (SKC-Single Stage; USA), containing blood agar culture medium at a sampling flow rate of 28.3 L/min. The tested filter media was fixed in a square-shaped holder (cross section area of 225 cm^{2} equivalent to the surface area, exposed to the filter air) and placed inside the test tunnel.

The air flow rate was adjusted in a way that the desired face velocity of the filter media (10 cm/s) was developed. Once the air sampling before and after test filter was done, the plates were kept inside 37^{0}C incubator for 48 h and the number of

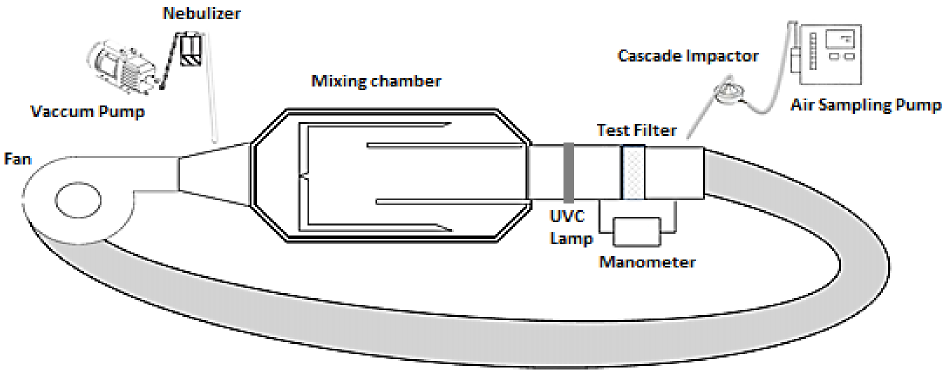


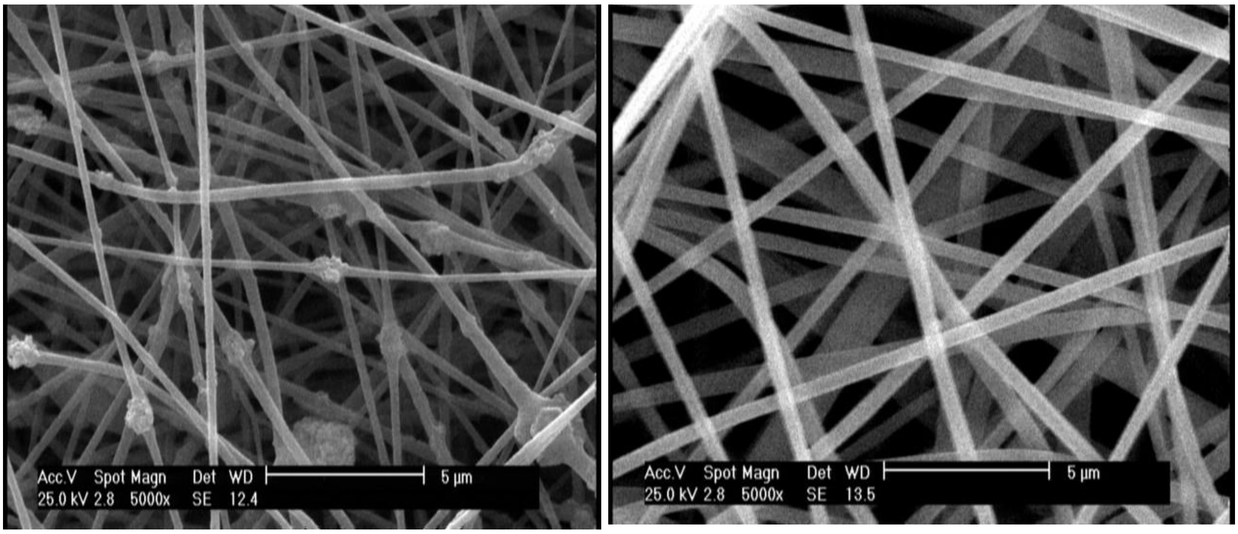
Fig. 1. Schematic design of filter test rig

colonies inside the culture medium was determined by colony counter (Schuett-Biotec GmbH; Germany). By calculating the difference between the number of upstream and downstream colonies of the filter, its efficiency percentage was calculated. Five samples of the studied filter media were prepared and tested. Afterwards, the mean percentage efficiency was calculated (ISO, 2011). It took 0.5 h for the sampling to be done, and the experiments were performed at 25^{0}C and relative humidity of 35±5%. In addition, the filters were also examined at two states: UVC radiation (Ushio; Japan) with an intensity of 1.8 mW/cm^2 (wavelength: 254 nm) and dark state. In Fig. 1, one can see the location of the blower fan to circulate the air in the tunnel test (in a closed cycle), the position of the nebulizer to spray the bacterial suspension with the help of vacuum pump before the mixing chamber, the place of the test filter and manometer for measuring the filter's pressure drop, and the cascade impactor to take air samples before and after the test filter (Mousavi et al., 2017).

The morphological studies of the nanofiber were carried out, using a scanning electron microscopy (SEM, Philips, XL 30; USA), after coating with gold. The diameter of the produced fiber was determined, using image processing software applications (Image J, National Institutes of Health; USA), and the porosity percentage of the nanofiber layer was determined with the help of algorithms to analyze SEM images via MATLAB software (MathWorks, Version 7), (SF Dehghan et al., 2016). The thickness of the nanofiber layer was determined, using a Caliper (ASIMETO- 307-56-3 6" ABS; Hong Kong), according to ISO29463-3:2011 recommendations (ISO, 2011). Fourier Transform Infrared Spectroscopy (FTIR) (Rayleigh- WQF-510; China) was performed on the hybrid nanofibers to detect its organic compounds and functional groups. Through wavelength-dispersive X-ray spectroscopy (WDX) in SEM microscope, the elemental detection of magnesium oxide nanoparticles in the nanofibers PAN/MgO was done. Furthermore, to reconfirm the presence of magnesium oxide nanoparticles in PAN/MgO nanofibers, X-ray diffraction spectroscopy (XRD) (STOE-STADIP' Germany) was also employed.

RESULTS AND DISCUSSION

Fig. 2 depicts SEM image with a magnification of 5000X from PAN/MgO nanofibers. The mean diameter of the PAN/MgO was 221.38±65.56 nm and 320.25±87.35 nm for PAN. Since the ratio of the standard deviation of the fiber diameter to the mean diameter fell below 0.3, the type of the produced fiber morphology was considered uniform (Matulevicius et al., 2014). The studied nanofibers showed a slight difference in the thickness (around 0.1 mm) and Grammage (around 17.5 g/m^2), while they had different conditions in terms of porosity (= packing density – 1). The porosity percentage of PAN/MgO and PAN was calculated as 41% and 51%, respectively.



PAN/MgO PAN

Fig. 2. SEM image of manufactured nanofibers

Results from assessment of nanofibers' morphological characteristics showed that by adding MgO nanoparticles to the polymer solution, the fiber diameter dropped while the number of beads rose. Studies have shown that addition of metal oxide nanoparticles to the polymer solution results in enhanced electric conductivity and, thus, elevated density of electric charge on the surface of polymer jet of the outflowed liquid. This causes greater jet stretching and lower diameter of the fiber during the electrospinning process (Dadvar et al., 2011). Here, PAN/MgO nanofibers had a lower fiber diameter and more beads, compared to PAN nanofibers. Under these conditions, it seems that the fibers had formed as something between electrospinning and electrospray states. MgO nanoparticles typically lie inside the PAN fiber, rather than the fiber surface. Accumulation of MgO powder can induce electro-spraying and bead formation. Nevertheless, more beads are observed in thinner fiber (Kim et al., 2012).

The porosity percentage of PAN/MgO nanofiber layer was lower than that of PAN one. It has been shown that by increasing the fiber diameter, the size of pores also enlarged. Bagherzadeh et al. (2013) indicated that lower concentrations of polycaprolactone electrospinning solution decreased fiber diameter, increased total volume of the fiber, and, thus, reduced porosity percentage (Bagherzadeh et al., 2013). Noorpoor et al. (2014) stated that with the reduction of electrospinning solution concentration, the fiber diameter shrank and the porosity percentage declined (Noorpoor et al., 2014). One study by Kwon et al (2005) concluded that the decrease of poly-L-lactide-co-caprolactone fiber's diameter, led to smaller porosity percentage, determined by the mercury intrusion porosimetry (Kwon et al., 2005).

Addition of MgO nanoparticles to PAN solution, could promote the removal efficiency of the bioaerosol, simultaneously increasing the pressure drop or air resistance of the filter. In terms of morphology, PAN/MgO nanofibers had more beads and less uniformity, compared to PAN ones. PAN/MgO layers had more packing density as well. These issues can boost filter resistance, lower air permeability, and raise pressure drop (Bao et al., 2016). Increased pressure drop of PAN/MgO filter, compared to PAN, can be a result of its lower fiber diameter. According to classic theory of filtration, pressure drop in continuous regimen has an inverse relationship with squared diameter of the fiber; however, for nanofibers, the increased level of pressure drop through reduction of the diameter has a smaller slope as a result of slip effect (Wang et al., 2008). Brown indicates that at a constant packing density of nanofibers, the increase in pressure drop could be observed with the decrease in fiber diameter even in the slip flow regime (Brown, 1993). It has been well proven that slip flow ($10^{-3}<$ Knudsen number (Kn) <0.25) occurs when air passes around the nanofibers. This is due to the fact that the fiber diameter is close to the mean free path of gas molecules (e.g., 65 nm for air under normal thermal and pressure conditions). In a slip flow, air speed is considered to be nonzero across the fiber (Hosseini and Tafreshi, 2010).

Fig. 3 illustrates the FTIR spectrum of PAN nanofibers. According to this figure, the vibrational properties of -C≡N group (the tensile nitrile group related to PAN) had emerged at 2241 cm^{-1}, while the peaks at 1221, 1363, and 1451 cm^{-1} were associated with the vibration of aliphatic groups (bending) of C-N (Yu et al., 2013). Mild stretching vibrations at the wavelength of 1600 cm^{-1} are related to C-C bond (Farsani et al., 2009).

Fig. 4 reveals the elemental map of magnesium oxide nanoparticles along with the image of PAN/MgO nanofiber (a), SEM image of PAN/MgO nanofiber (b), and elemental map of magnesium oxide nanoparticles (c). It has been done by WDX in SEM microscope for elemental detection of MgO nanoparticles in the PAN/MgO nanofiber, with the white points on the map indicating the presence of MgO particles in the studied sample.

Figure 5 illustrates XRD pattern of PAN/MgO nanofibrous filter, performed to reconfirm the presence of MgO nanoparticles. Three reflective peaks, corresponding to pure MgO crystal, emerging at $2\theta=36.8^0$, $2\theta=42.8^0$, and $2\theta=62.2^0$, suggest that the electrospun PAN/MgO nanofiber contained pure MgO crystals (Shao et al., 2006).

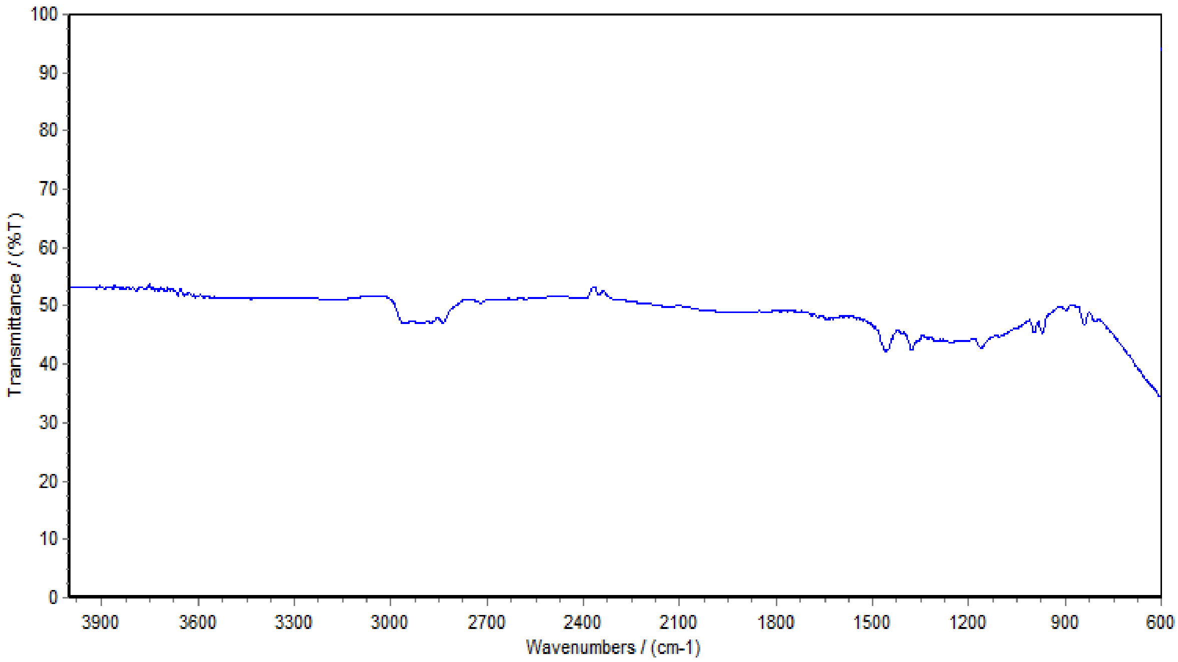
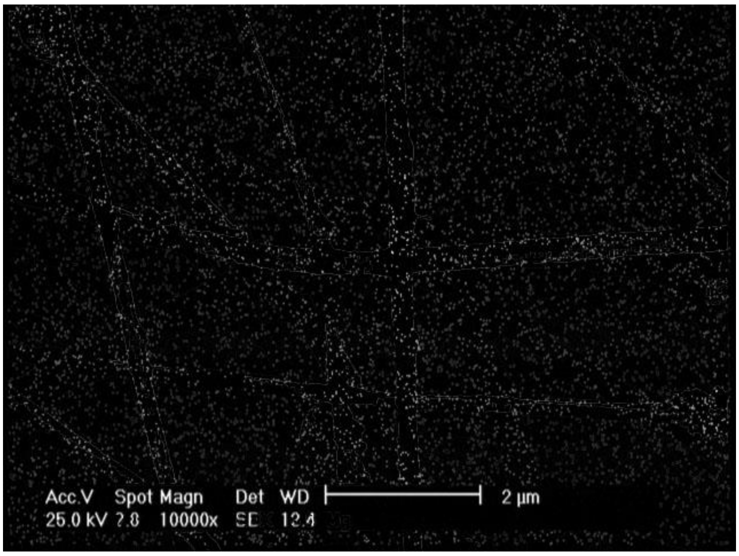
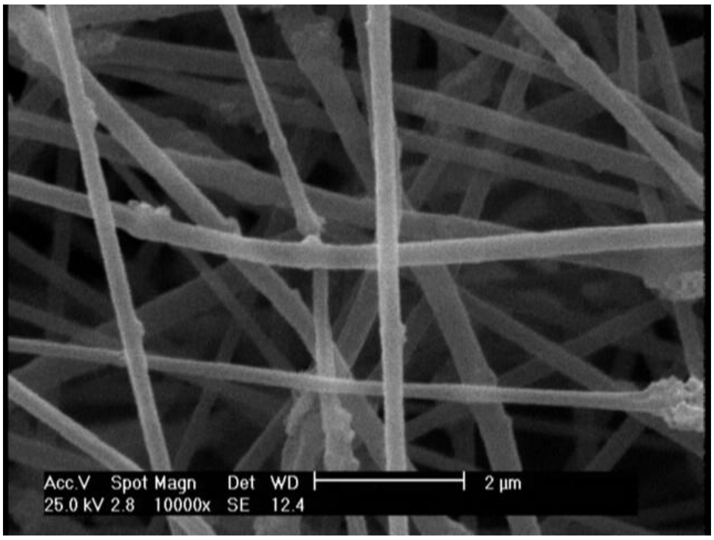


Fig. 3. FTIR spectra of PAN nanofibers

a



b



c

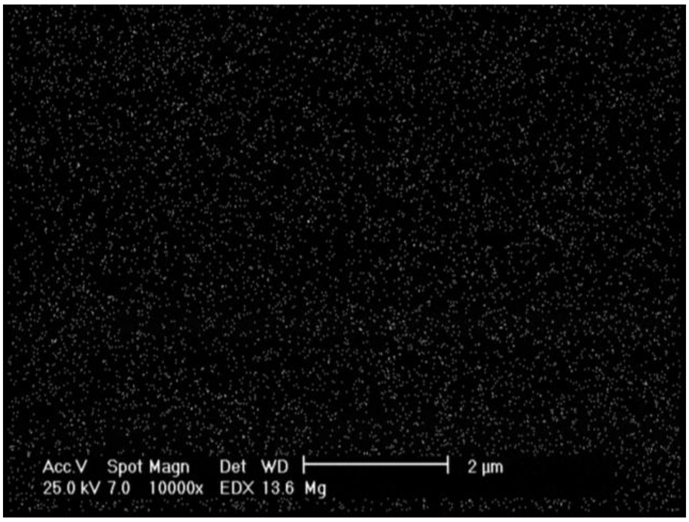


Fig. 4. Elemental map of MgO nanoparticles along with the image of PAN/MgO nanofiber (a), SEM image of PAN/MgO nanofiber (b), and elemental map of MgO nanoparticles (c)

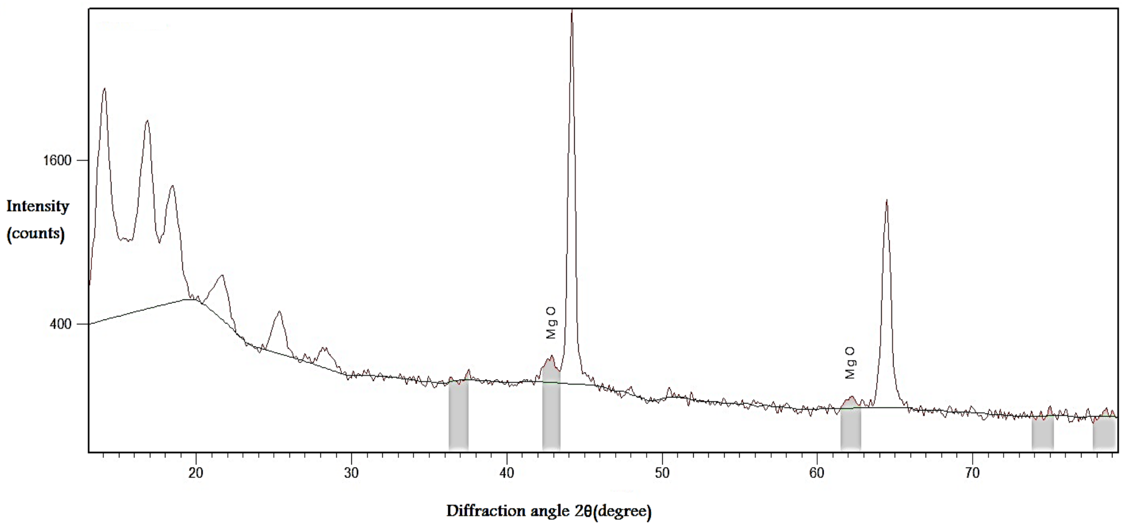


Fig. 5. X-ray diffraction Pattern of PAN/MgO nanofibrous filter

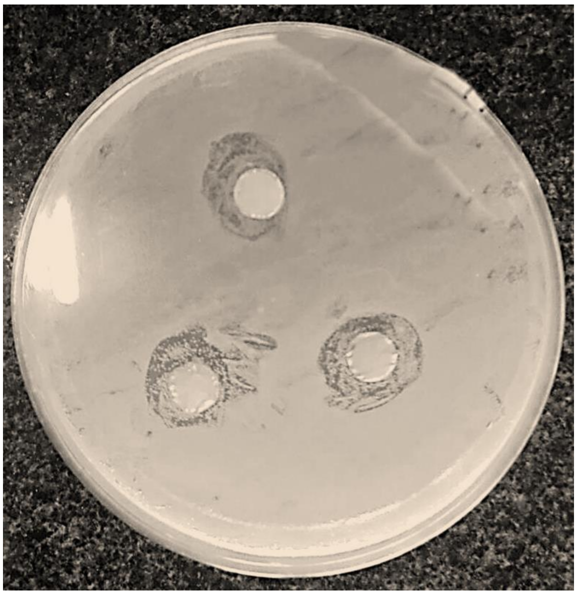


Fig. 6. Culture medium containing the blank discs

Results from the assessment of nanofibers' antibacterial properties, using disc diffusion method, confirm the antibacterial properties of PAN/MgO nanofiber. The mean length of the inhibition zone for PAN and PAN/MgO nanofibers turned out to be 0 and 2.8 mm, respectively. Fig. 6 illustrates the culture medium, containing blank discs of PAN/MgO nanofiber, along with a dark halo around the disc, which is a result of bacterial inhibition zone in these regions.

Table 1 provides the results of the mean removal efficiency of the studied nanofibers under two conditions, namely dark and UVC radiation. The mean percentage efficiency of filtration was higher in case of PAN/MgO, compared to PAN nanofiber filter; however, PAN/MgO nanofibers had higher mean pressure drop than the PAN ones. For both types of nanofibers under UVC radiation condition, the mean percentage efficiency for bioaerosol removal was higher than the dark condition.

Table 1. Removal efficiency of produced nanofibers under two conditions of dark and UVC radiation

Nanofiber Filter	Mean removal efficiency (%) UVC radiation	Dark	Mean pressure drop (pa)
PAN	85.27±15.63	52.15±11.43	120±28.71
PAN/MgO	99.98±5.24	82.50±14.35	296±45.18

Higher bioaerosol removal efficiency of PAN/MgO nanofiber layer can be attributed to its smaller diameter, greater packing density, and beaded morphology. According to classic theory of filtration, the efficiency has a direct relationship with filter thickness and fiber packing density, as well as an inverse relationship with fiber diameter and porosity (Wang et al., 2008). Fibers with smaller diameters possess higher surface area, higher packing density, and smaller pore size, thereby enhancing the filtration performance of the filter media. On the other hand, fibers with a larger diameter are typically bulkier and more porous, having a higher air permeability with lower pressure drop (Hutten, 2007). Wang et al. (2008) investigated the quality factor of filters, consisted of a nanofiber layer on a non-woven microfiber substrate. They concluded that the greater the density of nanofibers, the higher the efficiency and pressure drop. This is due to the increase in the solid component of the nanofiber layer, decrease in porosity and volume of pores, and thus increase in filtration area (Wang et al., 2008). Clearly, the removal efficiency grows as the fiber diameter decreases. This is in line with slip flow theory which says that the particles, suspended in the air, move closer to the fiber surface, whereby the chance of their captures is increased by interception (Papkov et al., 2013).

The results obtained from the test of mean difference of the bioaerosol concentration in upstream and downstream of the filter media indicated that the concentration of epidermidis microorganism under UVC radiation to the surface of both types of nanofibers decreased, compared to the dark condition. The effect of UVC radiation along with the use of MgO nanoparticles in reducing microorganisms' penetration has outperformed that of UVC radiation to the PAN nanofiber surface alone. It can be said that the performance of photocatalytic oxidation (MgO+UVC) has a significant effect on reduction of the extent of microorganisms' permeation, compared to the use of UVC alone in PAN nanofiber. Although UVC radiation is influential for reducing microorganisms' permeation in neat filters, the effectiveness of concurrent use of photocatalytic filters and UVC radiation has a more significant effect on decreasing the concentration of microorganisms in filter media's downstream. Thus, the performance of photocatalytic oxidation in the nanofiber was evaluated as a useful approach.

The study by Kühn et al. also suggested significant difference of epidermidis bacteria concentration at UV radiation condition, compared to no radiation. They stated that UV radiation can purify 80% of the bacteria, which is due to oxidative degradation of microorganisms by this radiation (Kühn et al., 2003). Chuaybamroong et al. used UV radiation to investigate the effect of photolysis on microorganisms' removal and found that the difference of bacillus subtilis concentration for typical HEPA filter during radiation and dark condition was statistically significant ($p=0.0001$) (Chuaybamroong et al., 2010). Zhang et al. (2010) employed PAN nanofiber (195 nm across), sandwiched between two active carbon fiber (ACF) mats, in a microwave-assisted way to remove aerosolized E. coli vegetative cells and B. subtilis endospores (Zhang et al., 2010) and concluded that in order to maximize the effects of microorganism disinfection on PAN nanofiber filters, microwave power should be the most important consideration (Zhang et al., 2010).

Several studies have been conducted to determine the effect of UV radiation on typical filter media to remove aerosol pathogens (Pigeot-Remy et al., 2014) (Lee et al., 2008); however, the present study examined the efficiency of nanofibers integrated with a photocatalytic material for removal of bioaerosols for the first time. The photocatalytic properties of MgO particles have been previously proven (Mageshwari and Sathyamoorthy, 2012). MgO nanoparticles show photocatalytic properties due to coral-like hierarchical structure, possessing large surface area along with porous nanoflakes network (Mageshwari and Sathyamoorthy, 2012).

Addition of MgO nanoparticles to nanofiber managed to enhance the filtration efficiency of epidermidis bioaerosol. It also increased the antibacterial properties, which is quite important in filtration. Since, air filters operate mostly under dark and humid conditions, they provide a suitable condition for the growth of bacteria, molds, and fungi (Lala et al., 2007). Presence of antimicrobial agents on the filter surface is considered a special advantage as it can boost the filtration efficiency. The antibacterial properties of MgO nanoparticles have been proven in several studies (Krishnamoorthy et al., 2012; Tang and Lv, 2014). The antibacterial activity of nanoparticles is due to their large surface area, abundance of crystal defects, and positively-charged particles, which can result in powerful interactions with negatively-charged bacteria and spores (Tang et al., 2012).

CONCLUSION

The present study aimed at synthesizing polymer nanofibers in two states, namely the neat one and based on magnesium oxide nanoparticles through electrospinning technique. It tried to investigate their filtration performance for bioaerosol removal from the air stream. The antibacterial properties of the manufactured nanofibers were also examined. Based on the obtained results, it can be stated that the produced PAN/MgO nanofibers can be useful for application in air conditioning units to purify the air coming indoors and also in local exhaust ventilation in industries or in fabric bag house. The produced nanofibers can be usefully applied in air conditioning units to purify the air coming indoors as well as in local exhaust ventilation in industries or in fabric bag house. Furthermore, one can use a nanofiber layer, containing MgO nanoparticles, along with the common commercial filters in air conditioning systems to increase their filtration performance.

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CONFLICT OF INTEREST

The authors declare that there is not any conflict of interests regarding the publication of this manuscript. In addition, the ethical issues, including plagiarism, informed consent, misconduct, data fabrication and/ or falsification, double publication and/or submission, and redundancy has been completely observed by the authors.

LIFE SCIENCE REPORTING

No life science threat was practiced in this research.

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