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Electrospun polyvinyl butyral/berberine membranes for antibacterial air filtration

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At the beginning of 2020, the whole world suffered from the new coronavirus (COVID-19). Wearing a mask was believed to reduce the spread of the virus. The core material of a mask required good air permeability and efficient filtration. Electrospun materials may match these requirements. By electrospinning, we prepared polyvinyl butyral (PVB)/berberine hydrochloride (BH) membranes onto the spunbonded nonwovens. The composite meshes showed porous structures, good air permeability (164 ± 16 mm/s) and air filtration efficiency 96.4% for PM 0.3, 100% for PM 2.5, with pressure drop (108 Pa). Moreover, with the addition of BH, the as-spun membranes showed good antibacterial property for staphylococcus aureus. Furthermore, the prepared PVB/berberine membranes had good hydrophobicity with water contact angle higher than 140°. These results indicated that the fabricated PVB/berberine membranes have potential applications in mask and air filtration.

1. Introduction

From the beginning of 2020, the world suffered from the new coronavirus (COVID-19). It was believed that wearing a mask may be a useful way to reduce the spread of the virus [1,2]. Generally, the filtration layer of a mask played the most important role to protect small droplet and particles partly due to its small pore size. The common filtration layer material was melt blown nonwoven fabric. However, during the outbreak of COVID-19, the output of the melt blown nonwoven fabric was shortfall. Several attempts were made to replace it [3–5]. Electrospun membrane was one of the choices due to its porous and controllable structures [6–8]. Moreover, during electrospinning process, kinds of additions could be blended into the as-spun fibers to form some special functions, for example antibacterial property [9–12].

Several research groups have reported the antibacterial electrospun membranes for air filtration. Ma et al. prepared electrospun polyvinyl alcohol (PVA)/chitosan (CS) and PVA/N-halamine nanofibrous membranes with antibacterial property for air filtration [13]. Bechelany et al. fabricated a novel silver/polyacrylonitrile (Ag/PAN) electrospun fibers used as air filters [14]. Kim et al. proposed a fabrication of antimicrobial nanofiber air filter using activated carbon and cinnamon essential oil (EO) blended into electrospun polyurethane fibers [15]. However, the antibacterial property of these electrospun membranes came from home-made materials, silver nanoparticles, or EO, which might be expensive for mass production.

Here, we reported a low-cost antibacterial electrospun membrane with the mixture of PVB and berberine hydrochloride (BH). We examined the effect of the concentration of berberine on the fabricated membranes including morphology, antibacterial property as well as the filtration efficiency. By the examination, we try to find an optimal formula to fabricate an economic and efficient electrospun membrane for air filtration.

2. Experimental

Polyvinyl butyral (PVB, Mw = 90000~120000 Da, Shanghai Aladdin Biochemical Technology Co., Ltd.) was dissolved in ethanol at 12 wt%, BH (Northeast Pharmaceutical Group Shenyang First Pharmaceutical Co., Ltd.) about 1.4 CNY/g pills were crushed and dissolved in the above solution at a ratio of 1 wt%, 2 wt%, and 5 wt%, and were denoted as BH1, BH2, BH5, respectively. The BH...
may be extracted from Coptis Chinensis, Phellodendron Amurense and berberis root plants, and its chemical structure was showed in Fig. 1. The prepared solution was stirred at room temperature for 8 h, and then let stand for 24 h before electrospinning. During the electrospinning process (Fig. 1), the prepared solution was loaded into a 5 mL syringe equipped with a 0.2 mm diameter stainless steel nozzle and then put into the electrospinning equipment (TTE-1, Qingdao Junada Technology Co., Ltd.). The voltage was set at 12 kV, the distance from the stainless steel nozzle to the receiver spunbonded nonwovens (SN, polypropylene, 13 g/m², Shandong Yuhang nonwoven Co., Ltd.) was 10 cm, and the solution feed rate was 0.3 mL/h.

The morphology, thickness, pore size, FTIR spectra of the prepared membranes were examined by a scanning electron microscope (SEM, Phenom Pro, Thermo Fisher Scientific), a handheld fabric thickness meter (YG141A, Wenzhou Jigao Testing Instrument Co., Ltd.), PSM 165 (Topas GmbH, Germany) and Nicolet iS10 spectrometer (Thermo Scientific), respectively. The air permeability was examined by a tester (Textest FX3300) under a pressure of 200 Pa in accordance with ASTM D 737. The water contact angle was tested by an optical tensiometer (Atttheta Theta, Biolin Scientific, Germany) within 3 loaded random sites with a 2 μL of deionized water drop. The filtration efficiency was determined by AFC 131 (Topas GmbH, AFC 131, Germany) maintaining flow rate 5.1 m³/min under solid sodium chloride (NaCl) aerosol from 3% NaCl solution. The antibacterial property was carried out by the agar plate diffusion method according to the previous study [10].

3. Results and discussion

Fig. 2(a-f) showed the morphologies of the SN, SN/PVB and SN/PVB/BH membranes loaded with BH at 1%, 2%, 5 wt%, and noted as SN/PVB/BH1, SN/PVB/BH2, SN/PVB/BH5, respectively. It can be
found in Fig. 2a that when the electrospun PVB fibers deposited onto the SN membranes, the larger pores in the SN (Fig. 2b) were covered and filled by the electrospun fibers and then formed a hierarchical structure. This structure was believed to be helpful for air filtration. As shown in Fig. 2d-f, with the addition of BH, the diameter of the electrospun fibers seemed to be increased firstly for BH1, BH2, while decreased for BH5, compared with pure PVB electrospun fibers (Fig. 2c). The water contact angles (WCA) of the prepared samples were shown in the inset images. As can be seen, the pure SN membranes showed superhydrophilicity with 0° WCA. Once the PVB was electrospun onto the SN membrane, the WCA increased obviously to about 140°. With the addition of BH, the WCA of the as-spun membranes did not change a lot, which may be due to the thinner fiber diameter and the rough surface of the membranes [16]. Moreover, the electrospun meshes only covered one side of the SN, which ensured that the other side still was hydrophilic allowing the exhaled water vapor transport. Once as the filtration layer of a mask, it could be beneficial to improve the one-way moisture conductivity of the mask and improve the comfort of wearing.

As shown in Fig. 3a, we found that the PVB/BH meshes had similar absorption peaks with pure PVB ones at wave numbers of 969 cm⁻¹, 1128 cm⁻¹, 2854 cm⁻¹ and 2957 cm⁻¹, which indicated that the addition of BH does not change the PVB chemical structure. In addition, the characteristic absorption peaks of the BH at 1370 cm⁻¹ and 2854 cm⁻¹ were also found in the PVB/BH samples, which indicated BH was blended into the PVB fibers. As displayed in Fig. 3(b)-(d), the pure SN, electrospun PVB and the SN/PVB composite membranes showed less antibacterial property against S. aureus. With the addition of BH, the composite membranes showed obvious inhibition zones (Fig. 3e-g). Moreover, it was found that the higher concentration of BH, the larger of the inhibition zones.

The filtration efficiency and the relative parameters of the samples were summarized in Table 1. It could be found that once electrospun fibers were loaded onto the SN meshes, the mean pore size of the composite membranes reduced sharply, and then the filtration efficiency increased obviously. The thicker of the electrospun meshes, the smaller pore sizes as well as the higher filtration efficiency. However, the filter pressure drop was also increased. For the PM 2.5, once the thickness of the electrospun membranes up to 0.01 mm, the filtration efficiency will reach 100%. For PM 0.3, it was harder to reach 100% efficiency with lower resistant. The optimal sample was the SN/PVB/BH5 with electrospun fibers thickness of 0.01 mm, and showed filtration efficiency of 96.4% for PM 0.3, 100% for PM 2.5, with filtration pressure drop 108 Pa. This efficiency will ensure the prepared composite membranes be a good choice for the filter layer of a mask.

4. Conclusions

In conclusion, we fabricated an antibacterial composite membrane with single side hydrophobicity and good air filtration efficiency through electrospinning process. This membrane contains a SN substrate and covered electrospun PVB/BH fibers. The optimal parameters of the as-spun fibers are BH concentration 5%, fiber diameter about 0.787 ± 0.222 μm, 0.01 mm thickness. The air filtration efficiency could reach 96.4% for PM 0.3 and 100% for PM 2.5 with a lower filter pressure drop 108 Pa. This membrane showed potential applications in mask and air filtration.

| Samples | Electrospun meshes thickness (mm) | Mean pore size (μm) | Gas permeability (mm/s) | Filtration efficiency PM 0.3% | Filtration efficiency PM 2.5% | Filtration pressure drop (Pa) |
|---------|----------------------------------|--------------------|------------------------|-----------------------------|-----------------------------|-----------------------------|
| SN      | 0.014                            | 204.4659           | 2820 ± 30              | 23.782                      | 19.881                      | 0                           |
| SN/PVB  | 0.009                            | 49.7272            | 722.33 ± 65.33         | 70.557                      | 95.003                      | 25                          |
| SN/PVB/BH1 | 0.010                      | 11.9240            | 451.17 ± 59.17         | 62.865                      | 97.57                       | 46                          |
| SN/PVB/BH2 | 0.015                      | 2.8356             | 124.4 ± 9.4            | 92.5                        | 100                         | 137                         |
| SN/PVB/BH5 | 0.024                      | 1.2966             | 108 ± 12.8             | 97.7                        | 100                         | 252                         |
| SN/PVB/BH1 | 0.007                      | 3.7902             | 186.83 ± 10.17         | 69.274                      | 98.916                      | 95                          |
| SN/PVB/BH2 | 0.011                      | 2.6566             | 175.5 ± 8.5            | 83.1                        | 100                         | 97                          |
| SN/PVB/BH5 | 0.024                      | 1.9291             | 88.1 ± 15.3            | 98.4                        | 100                         | 259                         |
| SN/PVB/BH1 | 0.007                      | 8.6971             | 355.67 ± 59.67         | 55.265                      | 98.93                       | 52                          |
| SN/PVB/BH2 | 0.010                      | 3.5712             | 104 ± 16               | 96.4                        | 100                         | 108                         |
| SN/PVB/BH5 | 0.011                      | 2.5349             | 155 ± 67.3             | 99.6                        | 100                         | 308                         |

Fig. 3. FTIR spectra of the samples (a) and antibacterial examination against S. aureus of SN membrane (b), electrospun PVB membrane (c), SN/PVB membrane (d), SN/PVB/BH1 (e), SN/PVB/BH2 (f), SN/PVB/BH5 (g).
**Declaration of Competing Interest**

The authors declare that they have no known competing financial interests or personal relationships that could have appeared to influence the work reported in this paper.

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