

SOUND ABSORPTION BEHAVIOR OF ELECTROSPUN POLYACRYLONITRILE NANOFIBROUS MEMBRANES*

Hai-fan Xiang^{a, b}, Shuai-xia Tan^a, Xiao-lan Yu^{a, b}, Yu-hua Long^{a, b}, Xiao-li Zhang^a,
Ning Zhao^a and Jian Xu^{a**}

^a *Beijing National Laboratory for Molecular Sciences, Laboratory of Polymer Physics and Chemistry, Institute of Chemistry, Chinese Academy of Sciences, Beijing 100190, China*

^b *Graduate University of the Chinese Academy of Sciences, Beijing 100049, China*

Abstract The acoustical damping property of electrospun polyacrylonitrile (PAN) nanofibrous membranes with different thicknesses and porosities was investigated. The sound absorption coefficients were measured using the impedance tube instrument based on ISO10534-2:1998(E). Results indicate that the first resonance absorption frequency of nanofibrous membranes shifts to the lower frequency with the increase of the back cavity or the thickness of membranes. Moreover, the sound absorption performance of the perforated panel can be greatly improved by combination with a thin layer of PAN nanofibrous membrane. Traditional acoustical damping materials (foam, fiber) coated with nanofibrous membranes have better acoustical performance in the low and medium frequency range than that of acoustical materials alone. All of the results demonstrate the PAN nanofibrous membrane is a suitable candidate for noise reduction.

Keywords: Polyacrylonitrile; Electrospinning; Sound absorption; Nanofibrous membrane.

INTRODUCTION

According to the report of World Health Organization (WHO)^[1], noise pollution has great adverse effects on human health such as noise-induced hearing impairment, disturbance of rest and sleep and so on. Therefore, developing excellent acoustical materials to prevent the noise pollution is an important issue. Traditional sound absorption materials include foams, fibers, membranes, perforated panels and so on. Among these acoustical materials, fibers are the most commonly used due to their good noise reduction performance. Researches on the fibrous sound absorption materials involve natural fibers^[2–4], metal fibers^[5], inorganic fibers^[6–8] and synthetic fibers^[9–11]. These traditional acoustical fibrous materials have good noise reduction abilities in the high frequency range, but exhibit little sound absorption properties in the low and medium frequency range (250–2000 Hz) in which human sensitivity to noise is high^[12]. Therefore, materials with excellent noise reduction properties in the low and medium frequency range are desirable in the acoustical field.

The diameter of the traditional acoustical fibers is primary in the range of micrometers. Different from microfibers, nanofibers have characteristics such as large specific surface area, high porosity, flexibility and extremely low weight. These outstanding properties are beneficial for obtaining preferable acoustical damping performance. For example, fibrous materials of combined nanofibers and microfibers have been manufactured recently^[13]. The combined materials show much better sound absorption behavior than that of the microfibers

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** Corresponding author: Jian Xu (徐坚), E-mail: jxu@iccas.ac.cn

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and can keep high sound absorption coefficients in both low and high frequency range. Consequently, nanofibrous materials are promising alternatives in the noise reduction field. However, there is few reports about nanofibrous membranes for acoustical application.

Electrospinning is a direct, simple and versatile method for producing nanofibers. The diameter of nanofibers and the physical structure properties (such as porosity, thickness and so on) of nanofibrous membranes can be easily tailored by adjusting the parameters of electrospinning. Different kinds of polymers are used for producing nanofibrous membranes by electrospinning. One of the most important materials is polyacrylonitrile (PAN), which has various applications due to its relatively cheap and good thermal, mechanical properties. Moreover, PAN fiber is also the most widely used precursor for carbon fiber. Therefore, electrospun PAN nanofibrous membranes, carbonized and activated PAN nanofibrous membranes have potential applications such as ultrafiltration membrane^[14], affinity membrane^[15], metal ions absorption^[16], hydrogen storages^[17], supercapacitors^[18], electrical conductive membrane^[19] and so on. However, few attempts have been made at preparing PAN nanofibrous membranes for the acoustical application and the relationship between the physical and acoustical characteristics of nanofibrous membranes has not been discussed enough and clarified.

Herein, PAN nanofibrous membranes were prepared by electrospinning and the sound absorption behavior of the nanofibrous membranes and their composites with traditional acoustical materials (perforated panel, foam and fiber) were evaluated. The results demonstrate that the nanofibrous membrane has a promising acoustical damping performance, especially in the low and medium frequency range.

EXPERIMENTAL

Materials

Polyacrylonitrile powders ($M_w = 7.38 \times 10^4$, PDI: 2.16) were provided by Shanghai Petrochemical Co., Ltd. Analytical grade *N,N*-dimethylformamide (DMF) was purchased from Beijing Chemical Reagent Plant. Commercial acoustical foam (thickness: 14 mm; apparent density: 0.01 g/cm³) was supplied by BASF Chemical Company. Polypropylene (PP) nonwoven (thickness: 3 mm; apparent density: 0.17 g/cm³) was produced by Huaфон Group of China. Perforated aluminous panel (thickness: 0.9 mm, porosity: 13%, diameter of pores: 1.5 mm) was supplied by Tsinghua University. Glass fiber (thickness: 15 mm, apparent density: 0.06 g/cm³) was purchased from Beijing Salable Environment Engineering Technologies Co., Ltd.

Sample Preparation

Polyacrylonitrile powders were dissolved in DMF at a concentration of 8 wt% at room temperature for 36 h. Electrospinning apparatus was set up horizontally and the applied voltage, tip-collector distance and flow rate were fixed at 10 kV, 13 cm and 0.02 mL/min, respectively. The electrospun nanofibrous membranes were collected on the aluminum foils. The temperature and the relative humidity were kept at $(17 \pm 2)^\circ\text{C}$ and $(60 \pm 10)\%$, respectively. In order to remove the residual solvent, the as-prepared PAN nanofibrous membranes were dried in an oven at 60°C for more than 8 h. The as-spun samples are 270, 298 and 325 cm² in size for electrospinning time of 10, 30, 60 min, respectively.

Test and Characterization

Morphology of the nanofibrous membranes was observed by FE-SEM (JEOL JSM-6700F). Fiber diameter and distribution were measured using image software ImageJ 2.0 (Statistical analysis of 50 nanofibers). The thickness of each nanofibrous membrane was the average value of ten measurements at different locations. The porosity (p) of electrospun nanofibrous membranes can be defined as follows:

$$p = (1 - \frac{\rho}{\rho_s}) \times 100$$

where ρ is the density of electrospun nanofibrous membranes (g/cm³), ρ_s is the density of PAN measured by densitometer (1.2329 g/cm³).

The rheological measurements were conducted with an Advanced Rheometric Expansion System (ARES,

Rheometric Scientific, NJ) which is a strain-controlled rheometer with a couette fixture. The temperature was controlled at $(17 \pm 2)^{\circ}\text{C}$. DDS-307 Conductivity Meter (Rex Electric Chemical Products Department, Shanghai Precision Scientific Instrument Co., Ltd) was used to measure the conductivity of PAN/DMF solution. The viscosity and the conductivity of the 8 wt% PAN/DMF solution are 1 Pa·s and 47.1 $\mu\text{S}/\text{cm}$, respectively.

Normal incidence absorption of electrospun nanofibrous membranes was measured by using Type SW 466 Impedance Tubes (BSWA Technology Co., Ltd., China) based on ISO10534-2:1998(E) (Acoustics-Determination of sound absorption coefficient and impedance in impedance tubes — Part 2: Transfer function method). The acoustical measurement system contains a tube, two microphones and a Frequency Analysis System MC3242 (Fig. 1a). The principle of testing normal incidence absorption of electrospun nanofibrous membranes is shown in Fig. 1(b). Broadband random sound waves are generated by the Loudspeaker at one end of the impedance tube, and then they transmit to the surface of sample at the other end. The reflected signals are picked up by the sensors. The sound absorption coefficient is defined as the ratio of the acoustical energy not be reflected by sample ($I_{\text{incident}} - I_{\text{reflected}}$) to the acoustical incident energy (I_{incident}). The large tube (60 mm diameter) can measure the sound absorption coefficients in the frequency range of 100–2500 Hz while the small one (30 mm diameter) can give the sound absorption coefficients in the frequency range of 1600–6300 Hz. The sound absorption coefficients in the full frequency range (100–6300 Hz) are the combination of the values measured in the two tubes. Each PAN nanofibrous membrane was tailored to desired circle pieces with a diameter of 3 cm and 6 cm for the acoustical test in small and large tubes, respectively (Fig. 1c). The nanofibrous membranes are vertically pasted in front of the sample holder part of the impedance tube (Fig. 1d). The depth of back cavity can be adjusted by the rigid plunger. All the acoustical tests were made at the temperature of $(17 \pm 2)^{\circ}\text{C}$ and relative humidity of $(60 \pm 10)\%$.

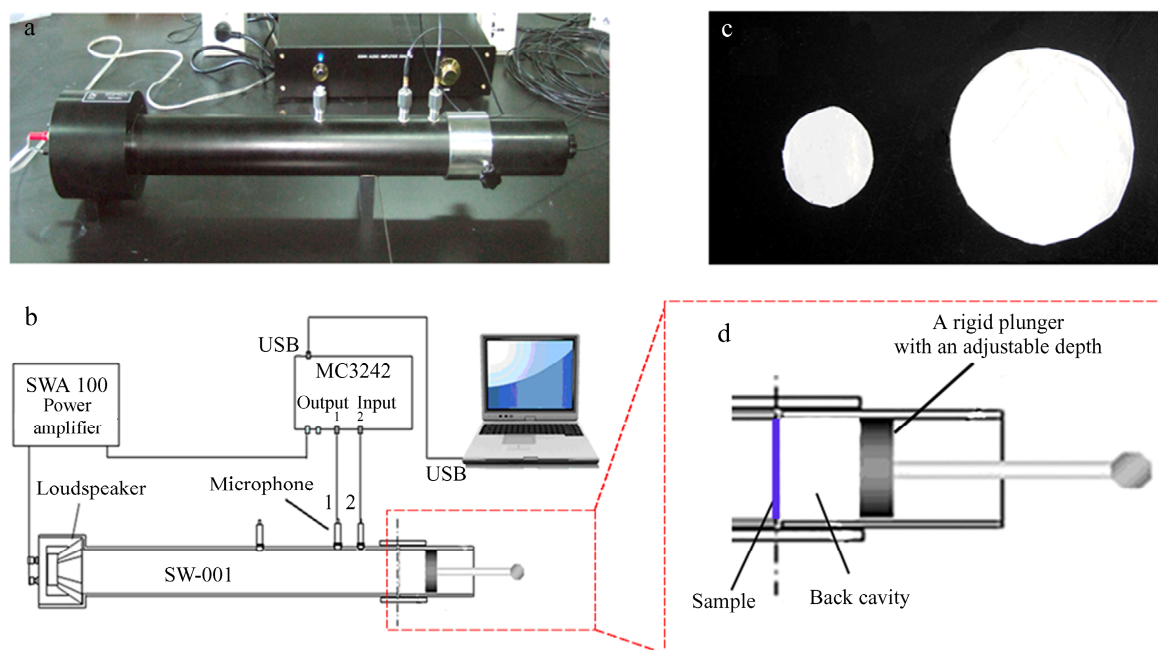


Fig. 1 (a) Photo of the impedance tube; (b) Schematic diagram of the acoustical measurement system; (c) Photo of the samples; (d) Schematic of the sample holder part of the impedance tube

The samples were test in three forms: (1) nanofibrous membranes with various back cavities; (2) nanofibrous membranes pasted on the perforated panel with certain back cavities; (3) nanofibrous membranes as coating layers on traditional acoustical materials.

RESULTS AND DISCUSSION

The Structure of Electrospun PAN Nanofibrous Membranes

As shown in Fig. 2(a), the average diameter of the as-spun nanofibers is about (333 ± 58) nm with a relatively narrow distribution. The thickness of PAN nanofibrous membranes increases from (17 ± 3) μm to (38 ± 3) μm and (205 ± 4) μm for the electrospinning time of 10, 30, 60 min (Figs. 2c–2e and Table 1). The samples are accordingly denoted as M1, M2, and M3. The porosity of M1, M2 and M3 is about 64%, 71% and 90%, respectively. The different porosities of samples can be attributed to the charges accumulated on the fibers which may increase with the electrospinning time, leading to a stronger repulsive interaction among the fibers and the enlarged spacing as a result.

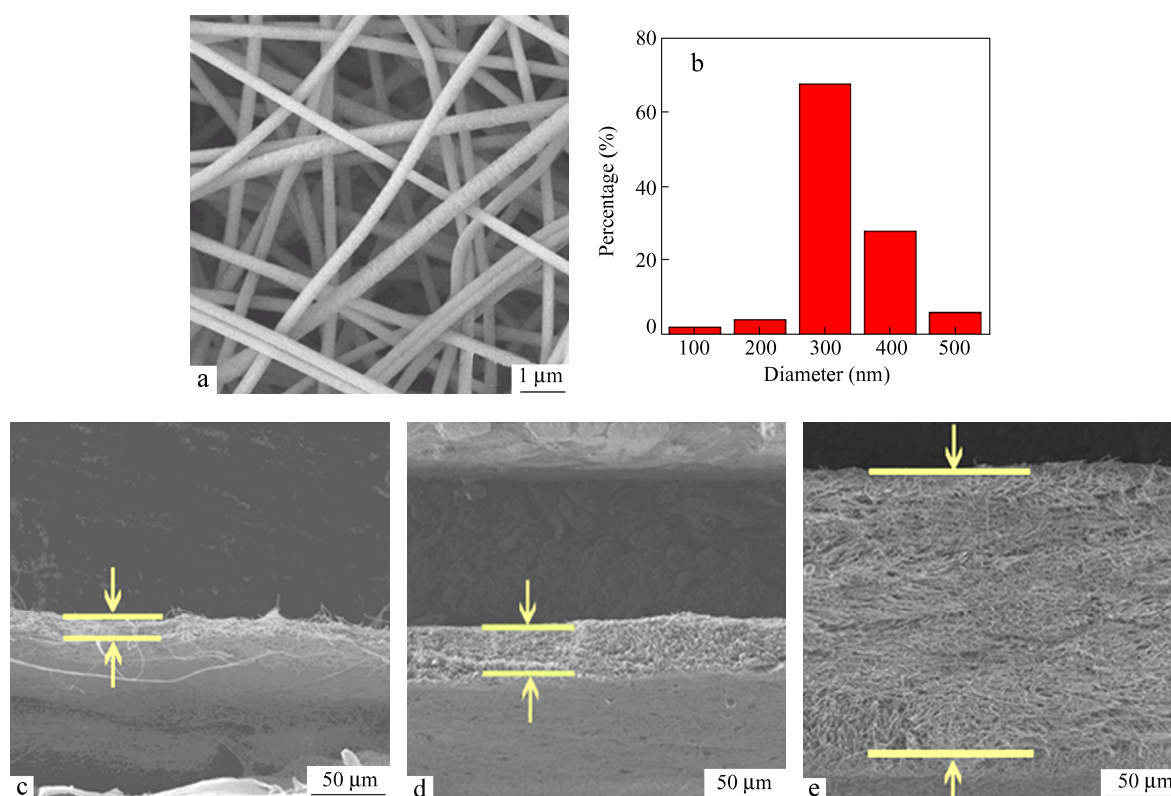


Fig. 2 SEM images of the PAN nanofibers (a), the diameter distribution of the nanofibers (b) and the profile of PAN nanofibrous membranes with different thicknesses tuned by electrospinning time: (c) 10 min, (d) 30 min, (e) 60 min

Table 1. Structure parameters of PAN nanofibrous membranes obtained from different electrospinning time

Sample No.	Electrospinning time (min)	Average thickness (μm)	Porosity, P (%)
M1	10	17 ± 3	64
M2	30	38 ± 3	71
M3	60	205 ± 4	90

Sound Absorption Property of Nanofibrous Membranes with Various Back Cavities

According to the acoustical theory, combining back cavities with thin fibrous materials is a usual method to improve the sound absorption performance in the low and medial frequency range for engineering applications since the effective thickness of the fibrous membranes can be increased by the back cavity^[20]. Moreover, this method is more economic than the traditional method in improving acoustical performance by increasing the thickness or buck density of the materials. However, until now, few reports concern the sound absorption

behavior of nanofibrous membranes with back cavities. Herein, we investigate the acoustical performance of nanofibrous membranes with different thicknesses and porosities at various back cavities (Fig. 3). The sound absorption coefficients of all the samples are below 0.1 without the back cavity (Fig. 3b) but dramatically increase and have a maximum value at a special frequency when tested with back cavities (Figs. 3c–3e).

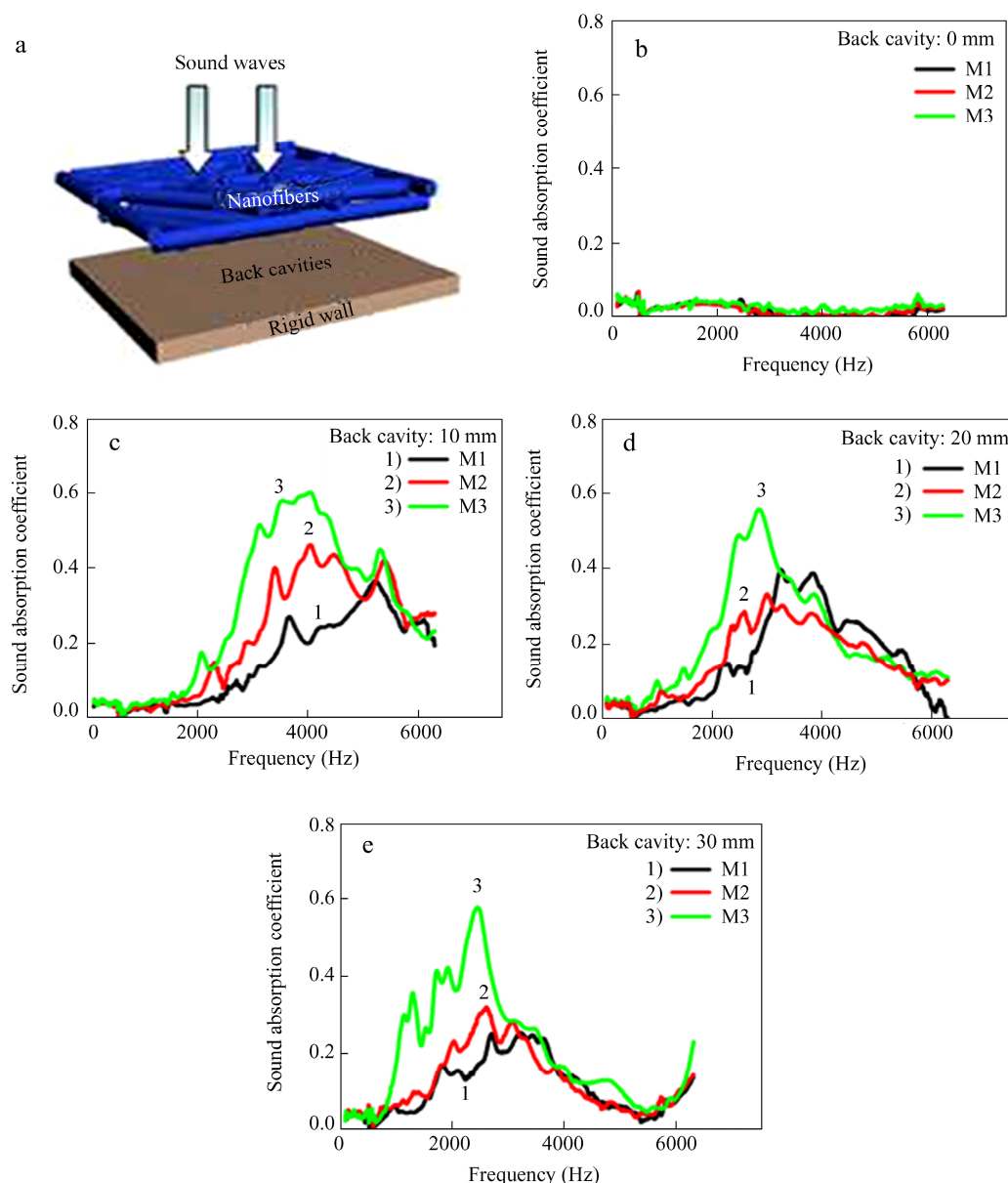


Fig. 3 (a) Schematic diagram of the testing form; (b–e) Sound absorption performance of nanofibrous membranes with various back cavities: (b) 0, (c) 10, (d) 20, (e) 30 mm

For example, the sound absorption coefficients of M3 with a back cavity of 10 mm are all above 0.1 in the frequency range of 2500–6300 Hz and have a maximum value of 0.6 at about 4000 Hz. The improvement can be explained as that the nanofibrous membrane and back cavity form Helmholtz resonators which absorb sound energy by the resonance of the system with various energy losses^[21]. Moreover, the first resonance frequency shifts to the lower frequency range with increasing the thickness of the membrane, *e.g.* the first resonance

frequency of M1 is 5154 Hz at the back cavity of 10 mm, while it decreases to 4247 Hz and 3854 Hz for M2 and M3, respectively (Fig. 3c). Besides, the maximum sound absorption coefficient also increases with the membrane thickness, varying from 0.37 for M1 to 0.46 for M2 and 0.6 for M3. This phenomenon of frequency-selecting sound absorption is also found in Figs. 3(d)–3(e) with other back cavities. In addition, the first resonance frequency also moves to the lower frequency range when the back cavities increase due to the inverse proportion between the resonance frequency and the back cavity. For instance, the first resonance frequency of M1 is 5154 Hz with a back cavity of 10 mm while it is 3248 Hz with a back cavity of 30 mm. Based on the investigation above, the sound absorption performance of nanofibrous membranes can be adjusted by tuning the thickness and porosity through the electrospinning process and the back cavities. It is worth to note that in our case, the thickness and porosity of PAN nanofibrous membranes are both dependent on electrospinning time. It is hard to get PAN nanofibrous membranes with different porosities but the same thickness. However, it will be interesting to investigate the effect of porosities of the nanofibrous membranes with a fixed thickness on their sound absorption property.

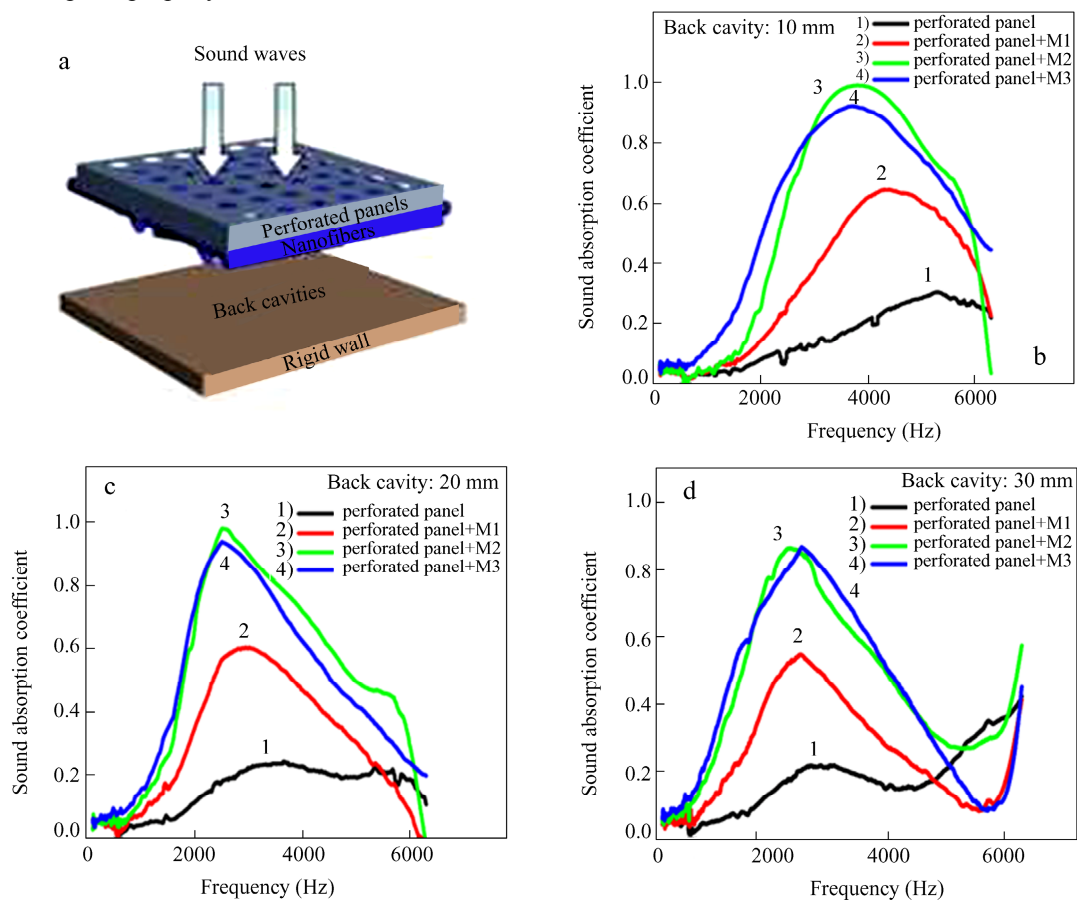


Fig. 4 (a) Schematic diagram of the testing form; (b–d) Sound absorption coefficients of perforated panel pasted with nanofibrous membranes with various back cavities: (b) 10, (c) 20, (d) 30 mm

Sound Absorption Property of Perforated Panel Pasted with Nanofibrous Membranes

The acoustical performance of perforated panel pasted with nanofibrous membranes has been evaluated (Fig. 4). Compared with the panel alone, the sound absorption coefficients of the composites with a certain back cavity greatly increase and the first resonance frequency moves to the lower frequency range (Figs. 4b–4d). Take Fig. 4(b) as an example, the maximum sound absorption coefficient of composites of M3 and the perforated panel with a back cavity of 10 mm is 0.93, which is three times higher than that of perforated panel alone,

indicating the additional thin layer of nanofibrous membrane can greatly enhance the acoustical property of the perforated panel. It can also be concluded from Figs. 4(b)–4(d) that the maximum sound absorption coefficients increase with the membrane thickness, but without significant difference when the thickness reaches a certain value. In addition, the first resonance frequency also moves to the lower frequency range as the back cavity increases.

Nanofibrous Membranes as High Resistance Coatings on Traditional Acoustical Materials

The sound absorption coefficients of other traditional acoustical materials have also been improved by employing a thin PAN nanofibrous membrane on the surface, as illustrated in Fig. 5. The sound absorption coefficients of the composites of BASF foam and PAN nanofibrous membranes are higher than that of BASF foam in a large frequency range unless a slight decrease in the high frequency and a similar tendency is also found for the glass fibers (Figs. 5b and 5c). For the PP nonwoven, the acoustical performance of the composite is improved in the whole frequency range (Fig. 5d). The improvement is very desirable for reducing noise in the daily life. The reason for the improvement may be attributed to the confined vibration of nanofibrous membranes on the traditional materials, resulting in more energy dispersion of the sound waves.

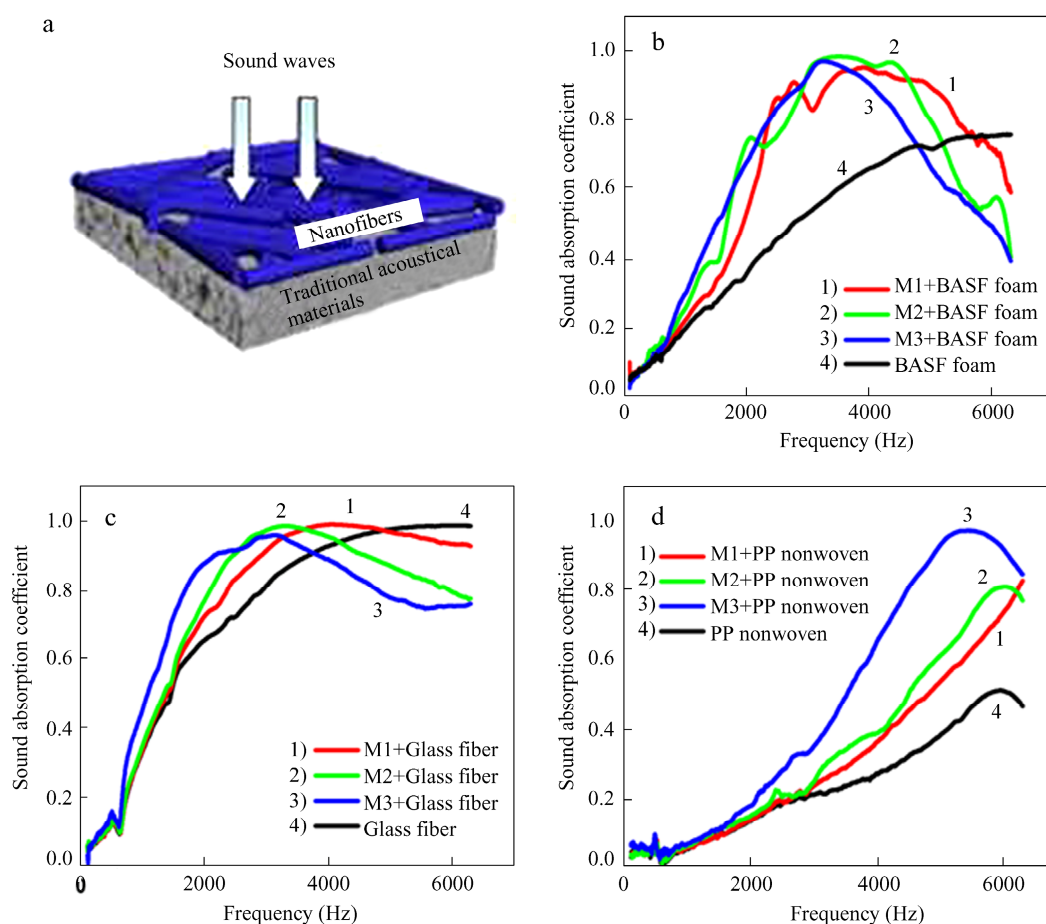


Fig. 5 (a) Schematic diagram of testing form; (b–d) Sound absorption properties of (b) BASF foam, (c) glass fiber and (d) PP nonwoven combining with PAN nanofibrous membranes without back cavities

CONCLUSIONS

In this paper, the sound absorption property of electrospun PAN nanofibrous membranes has been evaluated. Results exhibit that the first resonance frequency of the nanofibrous membrane shifts to the lower frequency

with the increase of the back cavity or the thickness of the membrane. The acoustical damping property of the resultant composites combining the nanofibrous membranes with traditional acoustical materials (perforated panel, foam and fiber) can be greatly enhanced especially in the low and medium frequency range, demonstrating the nanofibrous membrane a promising candidate for noise reduction.

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