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Improving Filtration Performance of Electrospun Nanofiber Mats by a Bimodal Method

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ABSTRACT: Nanofiber filtration is drawing an ever-increasing attention nowadays because of its high filtration efficiency as well as low basic weight. The objective of this study is to investigate the effect of structural characteristics on filtration performance with a single nanofiber mat between two pieces of nonwoven membranes. The filtration performance of nanofiber mats was evaluated by quality factor, the ratio of aerosol filtration efficiency to pressure drop. It was found that the quality factor dropped rapidly when the average fiber diameter (d_f) increased from 358 to 425 nm and decreased slowly from $d_f = 425$ nm to $d_f = 1250$ nm. This proved that gas-slip effect occurred on nanofibers with smaller diameters. Similarly, the quality factor of unimodal nanofiber mat declined as the packing density increased. Meanwhile, these data were compared with corresponding prediction of ideal mats predominantly from theoretical equations. Nanofiber mats with bimodal fiber size distributions were tested at the same condition. When compared with the unimodal nanofiber mats having the same weight-averaged fiber diameter and similar packing density, the bimodal nanofiber mats exhibited higher quality factors. Hence, the bimodal method is an effective method for the improvement of filtration performance. © 2012 Wiley Periodicals, Inc. J. Appl. Polym. Sci. 000: 000–000, 2012

KEYWORDS: filtration; nanofiber mats; bimodal; quality factor

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INTRODUCTION

Electrospun nanofibers were first introduced into industrial filtration about three decades ago by Donaldson using its proprietary Ultra-Web®.[1] However, the extensive investigations of electrospinning and nanofiber filtration were conducted in the past decade because of their unique properties such as high filtration efficiency and low basis weight.[2–6] Hajra et al.[7] discovered that the coalescence performance of the media improved largely when nanofibers were combined with the glass fiber media. Vitchuli et al.[8] found that depositing ultrathin Nylon 6 nanofiber mats on woven 50/50 nylon/cotton fabric can significantly improve the filtration efficiencies to 99.5% without sacrificing air permeability. Furthermore, Ahn et al.[9] prepared Nylon 6 nanofilter having the filtration efficiency of 99.993%, which was superior to the commercialized high efficiency particulate air (HEPA) filter. Actually, the basic weight of Nylon 6 nanofilter was only 10.75 g/m^2 when compared with 78.2 g/m^2 of the HEPA filter.

Nanofiber mats generally achieve an extremely high filtration efficiency with a relatively higher pressure drop across the nanofiber mats.[10] Quality factor, the ratio between aerosol filtration efficiency and pressure drop, can be used to evaluate the filtration performance of filters. Usually, larger value of quality factor indicates better quality of a filtration media. There are two primary ways to enhance the quality of an air filter: the first is to make it more efficient in filtering out aerosol to increase the filtration efficiency; and the second is to make it more permeable to reduce the pressure drop. Zhang et al.[11] reported that enhancing thickness uniformity of nanofiber mat was an efficient way to improve the quality factor. Yeom et al.[12] indicated that incorporation of additives with polymeric nanofibers was considered to be an important parameter to improve the quality factor by electrostatic charges. Yun et al.[13] further illustrated that the quality factor of beaded nanofiber mats was the best among various morphological structures (nanofiber, beaded nanofiber, and composite particle/nanofiber). Apparently, the morphological structure of nanofibers may play a pivotal role on the filtration performance.

Bimodal nanofiber mat is composed of a binary mixture of two fiber sizes. It is considered to have the potential to combine the benefit of thicker fibers, which span an open-pore structure as to keep the pressure drop low with properties of thinner fibers that are known to significantly enhance the aerosol filtration efficiency.[14,15] Nevertheless, to the best of our knowledge, no article has been published to discuss the quality factor of bimodal nanofiber mats.

The objective of this report is to investigate the effect of structural characteristics of polymer nanofibers on filtration performance. Polyacrylonitrile (PAN) used as a model was electrospun to prepare unimodal and bimodal nanofiber mats for filtration media. The filtration performance of the PAN nanofiber mats blended with polypropylene (PP) nonwoven membranes was evaluated on the basis of quality factor. Meanwhile, predictions from classical filtration theories were also checked against experimental results.

THEORETICAL ASPECTS

Fiber Packing Density

The packing density of nanofiber mat is given by the following equation:

$$\alpha = \frac{W}{\rho_f \times Z}, \tag{1}$$

where W is the basic weight of the nanofiber mat, ρ_f is the density of the polymer and has a value of 1.184 g/cm^3 for PAN, and Z is the thickness of the nanofiber mat.

Aerosol Filtration Efficiency

The aerosol filtration efficiency, η is defined as follows:

$$\eta = 1 - C_{\text{down}}/C_{\text{up}}, \tag{2}$$

where C_{up} and C_{down} are the aerosol concentrations before and after passing through the filtration media, respectively.

Aerosol filtration efficiency of a unimodal filtration media can be approximated by the following equation as described in the literature[16]:

$$\eta = 1 - \exp\left[-\frac{4\alpha\eta_f Z}{\pi(1-\alpha)d_f}\right], \tag{3}$$

where d_f is the average fiber diameter and η_f represents the total single-fiber efficiency.

The total single-fiber efficiency is the sum of single-fiber efficiencies due to interception, inertial impaction, diffusion, gravitational settling, and electrostatic attraction. For 300-nm particles, the single-fiber efficiencies due to diffusion and interception are dominant.[17] Total single-fiber efficiency is given as follows:

$$\eta_f = 1 - (1-\eta_D) \times (1-\eta_R), \tag{4}$$

where η_D and η_R are the single-fiber efficiencies due to diffusion and interception, respectively.

Different formulas are suggested for calculating single-fiber efficiencies due to diffusion and interception. Equations (5) and (6) present commonly used expressions[16]:

$$\eta_R = \frac{(1+R)^{-1} - (1+R) + 2(1+1.996Kn_f)(1+R)\ln(1+R)}{2(-0.75 - 0.5\ln\alpha) + 1.996Kn_f(-0.5 - \ln\alpha)}, \tag{5}$$

$$\eta_D = 2.27Ku^{-1/3}Pe^{-2/3}(1 + 0.62Kn_f Pe^{1/3}Ku^{-1/3}), \tag{6}$$

where $Ku = -\ln(\alpha)/2 - 3/4 + \alpha - \alpha^2/4$ is the Kuwabara hydrodynamic factor, $Pe = (U_0 d_f)/D$ is the Peclet number with U_0 as the face velocity, $D = K_B T C_S / 3\pi\mu d_p$ is the diffusion coefficient, K_B is the Boltzmann constant, T is the absolute temperature, μ is the air dynamic viscosity, d_p is the particle diameter, $R = d_p/d_f$ is the ratio of the particle diameter to the average fiber diameter, $Kn_f = 2\lambda/d_f$ is the fiber Knudsen number, and λ is the mean free path of the gas molecules (about 65 nm for air in normal temperature and pressure).

Pressure Drop

A unimodal filtration media's pressure drop is a function of air viscosity, thickness of filtration media, face velocity, average fiber diameter, and media's fiber packing density, which is given as follows:

$$\Delta P = f(\alpha) U_0 \mu Z / d_f^2. \tag{7}$$

Here, $f_{(\alpha)}$ is a function of fiber packing density only and has different forms based on different theories. The most popular correlation for calculating $f_{(\alpha)}$ is the empirical correlation,[16] given as follows:

$$f(\alpha) = 64\alpha^{3/2}(1 + 56\alpha^3). \tag{8}$$

Quality Factor

The quality factor, the ratio between aerosol filtration efficiency and pressure drop, is defined as follows:

$$\text{QF} = -\frac{\ln(1-\eta)}{\Delta P}. \tag{9}$$

A filtration media with greater filtration efficiency or lower pressure drop than another has higher quality factor. From eqs. (3) and (7), it is obvious to find that quality factor is independent of the thickness for a filtration media with unimodal fiber size distribution, in which α and d_f can be considered to be critical factors.

EXPERIMENTAL

Materials

PAN (MW = 150,000 g/mol) was received from Jiangsu Haide Group, Yancheng, China. *N*, *N*-Dimethyl formamide (DMF) was purchased from Aldrich. All chemicals and solvents were used without further purification. The PP nonwoven membranes (basic weight 22.6 g/m^2) with an average fiber diameter of about 30 μm were kindly provided by Nanjing Fiberglass Institute, Nanjing, China.

Preparation of PAN Nanofiber Mats

PAN was dissolved in DMF to prepare solutions with PAN concentrations of 6.5, 8, 9.5, 11, 12.5, and 14% (w/v, g/mL). A syringe pump was used to feed the polymer solution through a 20-mL plastic syringe fitted with a needle of tip diameter of 0.6 mm at a delivery rate of 4 mL/h. After high voltage ranging from 13 to 17 kV was applied to the needle, a positively charged jet of PAN solution was formed from the Taylor cone and sprayed to a grounded drum, ∼ 15 cm from the needle tip. With the evaporation of solvent, PAN nanofibers were deposited

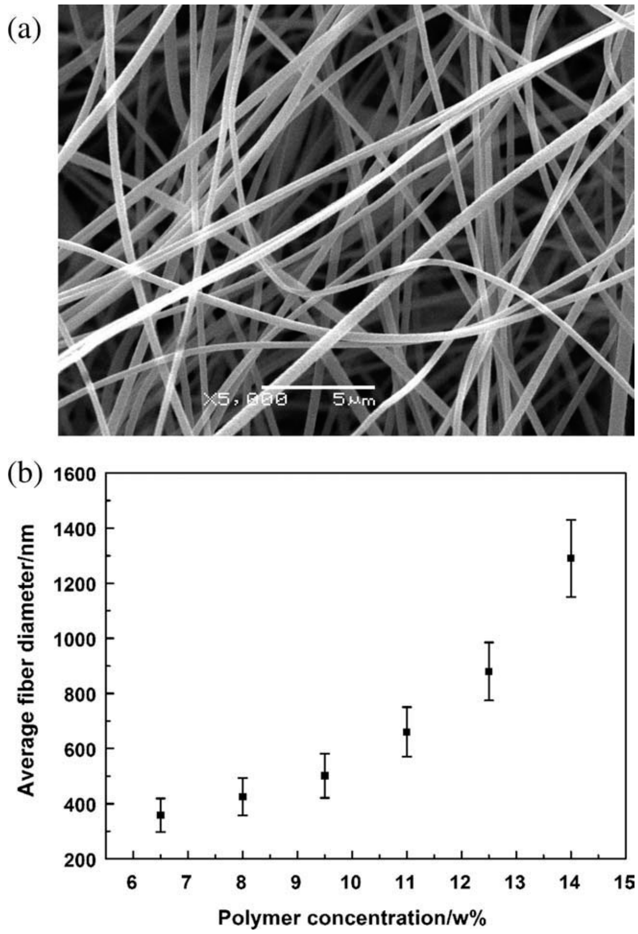

Figure 1. (a) SEM image of PAN mat composed of unimodal nanofibers with average diameter of 358 nm, and (b) the dependence of average fiber diameter on polymer concentration in spinning solution.

on the drum to form a nanofiber mat. Electrospinning experiments were carried out at 20°C and relative humidity of 50%. After the electrospinning was completed, the nanofiber mats were dried under vacuum and annealed at 80°C for 12 h to evaporate the excess solvent (DMF).

Nanofiber mats with bimodal fiber size distributions were prepared by the method as described previously in the literature[18] by using four different die/feeding systems in combination with two different polymer concentrations in the respective spinning solution. The weight fractions of the two types of fiber diameters in the mats were controlled through the feeding rates selected for the different dies.

Characterization

The morphologies of PAN nanofiber mats were observed with a scanning electron microscope (SEM; JEOL JSM-6360, Tokyo, Japan) after gold-sputter coating. The diameters of the electrospun nanofibers were measured directly from SEM images, with an average value being calculated from at least 100 measurements.

The packing density of the nanofiber mats was measured by the method as follows: samples were cut into squares with a length of 30 mm, and then, the thickness of the electrospun mats was determined from the combination of SEM cross-sectional images and micrometer. Finally, the packing density of the nanofiber mats is calculated by eq. (1).

Filtration Tests

Each electrospun PAN nanofiber mat was sandwiched between two pieces of PP nonwoven membranes as composite for filtration test. An automated filter tester (TSI 8130, Shoreview, MN) was used to measure the filtration efficiency and the pressure drop with dioctyl phthalate particle size of 300 nm and face velocity of 5.3 cm/s. The measurements were performed on the nominal filter area of 100 cm^2 at 20°C and relative humidity 25% in triplicate.

RESULTS AND DISCUSSION

PAN nanofiber mats were prepared by electrospinning polymer solutions with different concentrations to have a gradual increase in fiber diameters. SEM image of PAN nanofiber mat electrospun from PAN solution at 6.5 wt % is shown in Figure 1(a), clearly illustrating the acceptable uniformity for the filtration test in the following sections. Furthermore, the variation of the fiber diameter is shown in Figure 1(b). Obviously, the average diameter of the nanofibers increases from 358 to 1290 nm with increasing PAN solution concentration from 6.5 to 14% (w/v).

Fiber Packing Density

The packing density of PAN nanofiber mat was calculated using eq. (1) after determining the thickness for a given nanofiber mat as well as its basic weight. Figure 2 illustrates the packing density of the tested nanofiber mats (U_1–U_6) as a function of the average fiber diameter, where the basic weight is kept constant. The packing density of U_1–U_6 turned out to be about 0.057–0.067, independent of the average fiber diameter considered. The packing densities of U_1 ($W = 0.788$ g/m^2), U_7 ($W = 1.483$ g/m^2), U_8 ($W = 2.442$ g/m^2), and U_9 ($W = 3.895$ g/m^2) are compared in Figure 3. These four samples had the same average fiber diameter ($d_f = 358$ nm) and were tested under the same conditions. Obviously, the increase of the basic weight from 0.788 to 3.895 g/m^2 elevates the fiber packing density

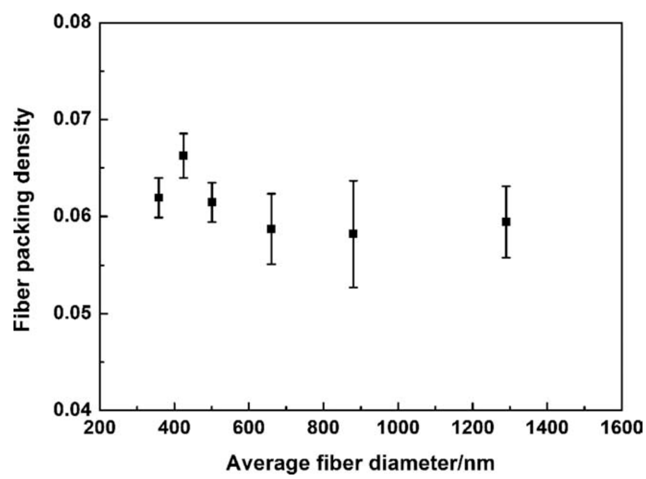

Figure 2. Fiber packing density against average fiber diameter of the unimodal PAN nanofiber mat.

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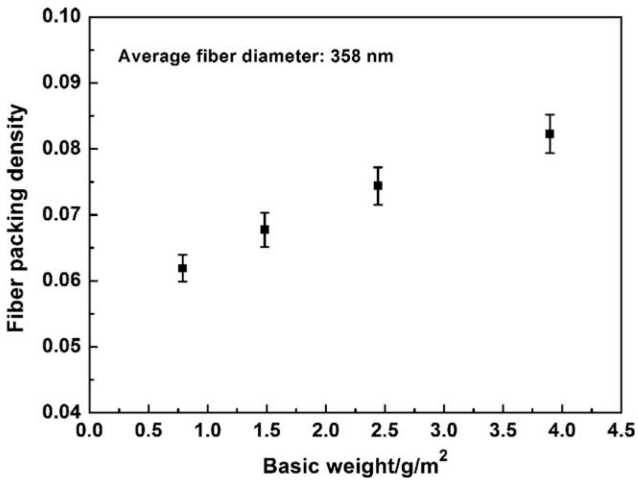

Figure 3. Fiber packing density against basic weight of the unimodal PAN nanofiber mat.

from 0.0619 to 0.0823. This result also confirms the argument in the literature.[19]

Background Collection

A nanofiber mat was too thin to have the mechanical strength for filtration tests,[20] and therefore, a supporting structure with sufficient mechanical strength was demanded. The substrate has been taken as two extremely permeable two pieces of PP nonwoven membranes with limited filtration efficiency so that the filtration efficiency and the pressure drop actually across the nanofiber mat can be calculated precisely.

To evaluate the practical performance of the PAN nanofiber mats only, both the pressure drop and the aerosol filtration efficiency of the substrate should be eliminated from the data of the composites. Their aerosol filtration efficiency and pressure drop are given by eqs. (10) and (11).[11]

$$\eta_N = 1 - (1 - \eta_C)/(1 - \eta_S), \qquad (10)$$

where η_C is the aerosol filtration efficiency of the composite, and η_N and η_S are the aerosol filtration efficiency of the nanofiber mat and the substrate, respectively.

$$\Delta P_N = \Delta P_C - \Delta P_S, \qquad (11)$$

where ΔP_C is the pressure drop of the composite, and ΔP_N and ΔP_S are the pressure drop of the nanofiber mat and the substrate, respectively.

Nanofiber Mats with Unimodal Diameter Size Distribution

Quality Factor Versus Average Fiber Diameter. The filtration testing results of unimodal nanofiber mats are presented in Table I. The aerosol filtration efficiencies of the 300-nm aerosol through the U_1, U_2, U_3, U_4, U_5, and U_6 were 91.29, 82.1, 66.7, 42.3, 23.3, and 12.1%, respectively. As shown in Table I, aerosol filtration efficiency of unimodal nanofiber mat obviously decreases with respect to increase in average fiber diameter from 358 to 1290 nm. Likewise, the pressure drop across the unimodal nanofiber mats with different fiber diameters shows a corresponding trend.

Advanced filtration filter gives high aerosol filtration efficiency and low pressure drop, thus larger value of quality factor indicates better quality of a filtration media. Figure 4 shows the results for the quality factor as a function of the average fiber diameter of unimodal nanofiber mats. As described in Figure 2, six samples have a similar fiber packing density. The theoretical quality factor of unimodal nanofiber mat can be obtained by calculating eqs. (3)–(9). In Figure 4, the red line corresponding to theoretical quality factors of U_1–U_6 shows a generally decreasing trend against average fiber diameter. Experimentally, when the average diameter of the nanofiber mat is 358 nm, the quality factor of this nanofiber mat is shown to be 0.0375 Pa^{-1}. However, as the average diameter of the nanofiber mat increases to 1290 nm, the quality factor decreases to 0.0236 Pa^{-1}. The experimental data also agree with this trend. When compared with the theoretical results, the quality factor decreases more sharply when average fiber diameter increases from 358 to 425 nm in experiment. This result supports that gas-slip effect occurs on nanofiber mats with diameters smaller than 500 nm.[1]

Quality Factor Versus Fiber Packing Density. Table I shows the filtration efficiency of nanofiber mats U_1, U_7, U_8, and U_9 under face velocity of 5.3 cm/s. The packing density of nanofiber mats increases from U_1 ($\alpha = 0.0619$) to U_9 ($\alpha = 0.0823$).

Table I. Properties of Unimodal PAN Nanofiber Mats and Substrate

Sample	Average fiber diameter (nm)	Basic weight (g/m^2)	Thickness (μm)	Aerosol filtration efficiency (%)	Pressure drop (Pa)
Substrate	30,000	22.6 × 2	(152.5 ± 3.5) × 2	4.8	3
U_1	358	0.788	10.75 ± 0.35	91.29	65
U_2	425	0.804	10.25 ± 0.35	82.1	50
U_3	501	0.782	10.75 ± 0.35	66.7	34
U_4	660	0.798	11.5 ± 0.71	42.3	19
U_5	880	0.772	11.25 ± 1.06	23.2	10
U_6	1290	0.808	11.5 ± 0.71	11.1	5
U_7	358	1.483	18.5 ± 0.71	98.92	128
U_8	358	2.442	27.75 ± 1.06	99.936	220
U_9	358	3.895	40.00 ± 1.41	99.999	365

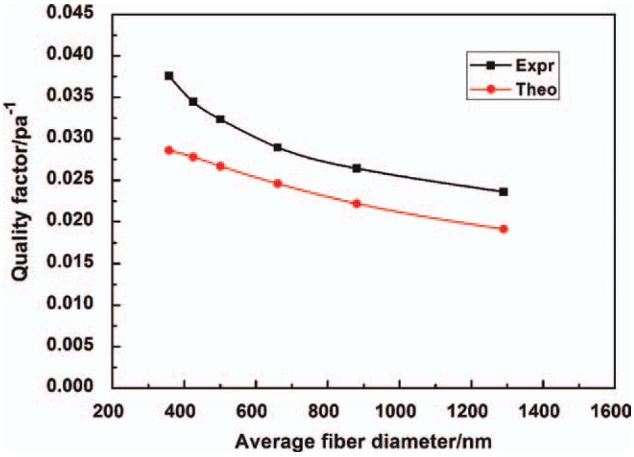

Figure 4. Quality factor as a function of average fiber diameter of the unimodal PAN nanofiber mat. [Color figure can be viewed in the online issue, which is available at wileyonlinelibrary.com.]

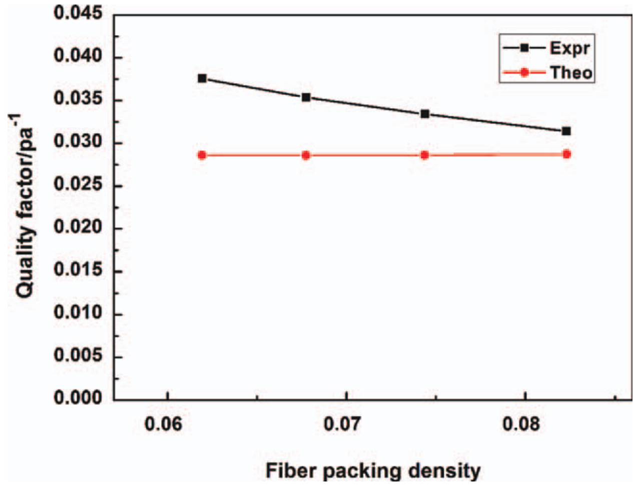

Figure 5. Quality factor as a function of fiber packing density of the unimodal PAN nanofiber mat having the average diameter of 358 nm. [Color figure can be viewed in the online issue, which is available at wileyonlinelibrary.com.]

It follows that filtration efficiency increases with fiber packing density. The pressure drops of the nanofiber mats with different fiber packing densities were also investigated. It was found that the pressure drop increased from 65 to 365 Pa when the fiber packing density increased from 0.0619 to 0.0823.

Figure 5 shows the dependence of quality factor on the nanofiber packing density under face velocity of 5.3 cm/s at the selected aerosol size. The result illustrates that quality factor drops steadily when fiber packing density increases from 0.0619 to 0.0823 g/m^2. Hence, it is desirable to use the smallest packing density of nanofiber mats as possible in order to maximize the quality factor. As theoretical model is based on hypothesis, the theoretical result does not agree with the measured trend. Similar experimental results were also reported by another group.[20]

Nanofiber Mats with Bimodal Diameter Size Distributions

Bimodal nanofiber mats were prepared by using two different die/feeding systems in combination with two different polymer concentrations in the respective spinning solution. Three pairs of the PAN spinning solutions combination were chosen whose concentrations were as follows: 6.5 and 14%, 8 and 12.5%, and 9.5 and 11% (w/v) in DMF. The weight ratios of the two types of fiber diameters in the resulting nanofiber mats were controlled through the feeding rates; more details are given in Table II. Figure 6 shows SEM image of PAN nanofiber mat characterized by a bimodal fiber diameter distribution, composed of thicker nanofibers (1290 nm) and thinner nanofibers (358 nm).

The weight ratio of the two different nanofibers and their average fiber diameters were used to define an effective fiber diameter: weight-averaged diameter. This value is largely used for comparison of the data with those obtained for unimodal nanofiber mats.[21] Meanwhile, through controlling the basic weigh of bimodal nanofiber mats, the fiber packing density of these mats turned out to correspond to the one found for unimodal nanofiber mats, a value between 0.057 and 0.067 was found.

Table II. Properties of Bimodal PAN Nanofiber Mats

Sample	Feeding rate of lower PAN concentration (mL/h)	Feeding rate of higher PAN concentration (mL/h)	Thickness (μm)	Weight-averaged fiber diameter (nm)	Basic weight (g/m^2)	Aerosol filtration efficiency (%)	Pressure drop (Pa)
B_1	2	2	11.25 ± 0.35	824	0.808 ± 0.033	85.4 ± 0.7	55 ± 2
B_2	1.3	2.7	11.00 ± 0.35	979	0.804 ± 0.029	76.4 ± 1.9	44 ± 2
B_3	1	3	10.75 ± 0.71	1057	0.792 ± 0.010	62.7 ± 2.1	32 ± 2
B_4	2	2	11.00 ± 0.71	653	0.786 ± 0.062	77.8 ± 1.8	47 ± 4
B_5	1.3	2.7	11.75 ± 0.35	728	0.820 ± 0.076	65.1 ± 3.2	34 ± 1
B_6	1	3	11.5 ± 1.06	766	0.815 ± 0.023	54.2 ± 1.3	26 ± 1
B_7	2	2	11.25 ± 0.35	581	0.810 ± 0.039	62.6 ± 2.1	31 ± 2
B_8	1.3	2.7	11.00 ± 0.35	607	0.783 ± 0.041	54.9 ± 1.3	27 ± 1
B_9	1	3	11.5 ± 0.71	620	0.792 ± 0.039	51.9 ± 2.5	25 ± 2

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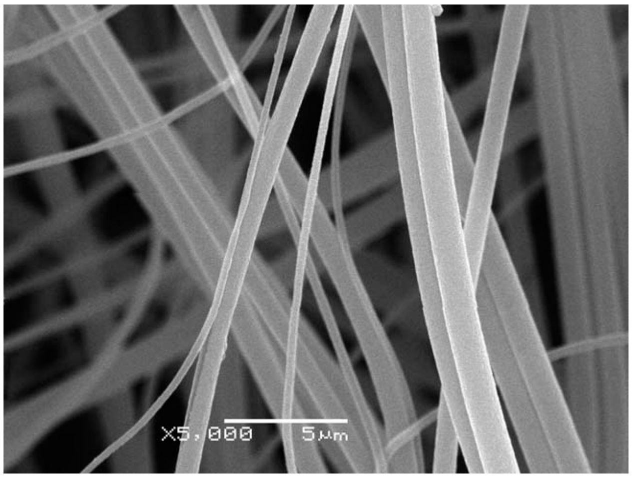

Figure 6. SEM image of PAN mat composed of bimodal nanofibers with the average diameters of thicker fibers and thinner ones amount to 1290 nm and 358 nm, and the weight ratio of thinner fibers in the whole mat is 1:3.

The filtration tests on the bimodal nanofiber mats are given in Table II. An interesting observation for quality factors of bimodal nanofiber mats as obtained by calculating the data from Table II is obvious from Figure 7. When compared with the unimodal nanofiber mats having the same weight-averaged diameter together with similar packing density, the bimodal nanofiber mats have higher quality factors. More interestingly, the bimodal nanofiber mats constructed from two most dissimilar fiber diameters show the highest quality factors. This is largely because the bimodal nanofiber mats combine the benefit of thicker fibers, which reduce the pressure drop with properties of thinner fibers that are known to increase the aerosol filtration efficiency. In applications that require high quality factor, it is more effective to use bimodal methodology to prepare a nanofiber mat instead of using a unimodal nanofiber filter.

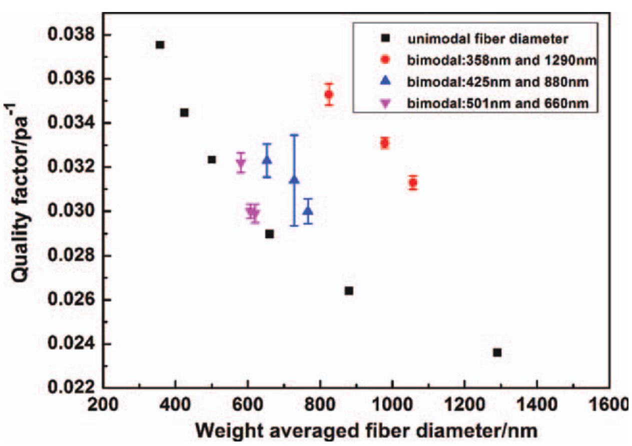

Figure 7. Quality factor of unimodal PAN nanofiber mat and bimodal PAN nanofiber mat as a function of weight-averaged fiber diameter. [Color figure can be viewed in the online issue, which is available at wileyonlinelibrary.com.]

CONCLUSIONS

PAN nanofiber mats were prepared by electrospinning for application to a filtration media. The effect of average diameter together with packing density of unimodal nanofiber mats on filtration performance has been investigated. The decrease in both the average diameter and the packing density shows the improvement of the quality factor of unimodal nanofiber mats. When compared with the unimodal nanofiber mats having the same weight-averaged diameter and similar packing density, bimodal nanofiber mats showed higher quality factors. Therefore, the bimodal method should be an effective way to develop high-performance nanofiber filtration systems.

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