Research Article

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Theoretical and experimental studies on the fabrication of cylindrical-electrode-assisted solution blowing spinning nanofibers

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Abstract: Cylindrical-electrode-assisted solution blowing spinning (CSBS) is a novel technique of fabricating nanofibers. In this paper, a combination of numerical simulation, theoretical analysis, and experiment is used to study the influences of CSBS airflow field and electric field on the fabrication of CSBS nanofibers for the first time. The effects of air pressure and injection speed on the morphology of CSBS fiber are studied. The research results show that the increase in air pressure will increase the centerline velocity and the centerline turbulence intensity within the effective stretching distance of the airflow. The increase in centerline velocity will result in a decrease in the diameter of CSBS fibers. There is a negative correlation between jet diameter and surface charge density of CSBS jet. The increase in air pressure will increase the stretching of the jet by the air flow, which will make the jet more likely to become thinner again because of the charge repulsion. Increasing air pressure will reduce the porosity of the nonwoven. As the injection speed increases, the diameter of CSBS fiber increases, and the porosity of the nonwoven decreases first and then increases. This work provides theoretical and experimental bases for the controllable preparation of CSBS nanofibers.

Keywords: cylindrical-electrode-assisted solution blowing spinning, nanofibers, effective stretching distance, surface charge density, morphology

1 Introduction

As one of the academic hotspots in the world, the research on nanofibers has developed rapidly in recent years. The ultrafinesness of the radial dimension gives nanofibers such excellent properties as size effect and surface effect that are different from conventional fibers. Nanofibers are therefore widely used in biomedical application (1–5), packaging materials (6), air filtration application (7–9), energy application (10–13), and many other fields. Traditional methods for fabricating nanofibers include electrospinning (ES) (14–16), solution blowing spinning (SBS) (17,18), phase inversion (19), centrifugal spinning (20), template synthesis (21), etc. Among them, SBS is one of the nanofiber preparation methods with the most large-scale and industrialized production potential (17,22). However, the nanofibers fabricated by the SBS have some defects such as large diameter and large standard deviation of diameter (23,24). These defects not only reduce the quality of SBS nanofibers but also limit the application of SBS nanofibers. A novel nanofiber fabrication method called cylindrical-electrode-assisted solution blowing spinning (CSBS) was developed in our previous research. Different from the traditional SBS, the CSBS equipment has added a cylindrical-assisted electrode. CSBS can effectively improve the quality of nanofibers. Compared with SBS nanofibers, the standard deviation of the diameter of CSBS nanofibers can be reduced by 21%, and the average diameter can be reduced by 6.17% (25). The CSBS jet is attenuated under the combination of air-flow stretching and electric field force, which makes CSBS different from traditional nanofiber preparation methods in terms of fabrication mechanism and equipment structure. Although there have been some theoretical and experimental studies on the air flow field (SBS (26) or melt blown (27,28)) and the electric field (ES (29,30)) in some nanofiber preparation systems (31), the complexity of CSBS has led to the lack of research that considers both the airflow field and electric field on the preparation of

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CSBS nanofibers. Insufficient research on the preparation mechanism and fabrication effect has hindered the promotion and application of CSBS technique.

In this paper, the finite element simulation software (ANSYS 2020 R2) is used to simulate the CSBS airflow field, and the distribution of the CSBS airflow field is studied. The influences of air pressure on the centerline velocity and the centerline turbulence intensity within the effective stretching distance are discussed. The effect of air flow field and electric field on the fabrication and morphology (fiber diameter and nonwoven porosity) of CSBS fibers under different air pressures are studied by theoretical analysis and experiments. In addition, the relationship between injection speed and CSBS fiber morphology is studied in this work.

2 Experimental

2.1 Materials

Poly(ethylene oxide) (PEO) ($M_w = 10^6$ g/mol) was purchased from Shanghai Liansheng Chemical Co., Ltd. (China). 7 g of PEO powder was added to 93 g of self-made distilled water and stirred with a magnetic stirrer (X85-2S, Meiyongpu Instrument Co., Ltd., China) at room temperature for 12 h to prepare 7 wt% PEO solution.

2.2 Equipment and process

The schematic of the CSBS setup is shown in Figure 1. The air compressor (A) generated high-pressure gas and delivered the gas to the die (B). The micro-syringe pump (C) pushed the polymer solution through the teflon tubing (E) to the blunt needle (length = 38 mm, inner diameter = 0.42 mm, outer diameter = 0.72 mm) (F). The tip of the blunt needle was facing the center of the die's outlet. The solution extruded from the needle was stretched into a jet by the high-speed airflow ejected from the die. The jet flew through the cylindrical-assisted electrode (G) and then formed nanofibers on the receiver (I). The electrode (G) was connected to the high-voltage power supply (H). The CSBS jet will be charged with the same kind charges because of electrostatic induction in the electric field formed by the electrode. When the charge repulsion is greater than the surface tension of the jet, the jet will split

Figure 1: Schematic of CSBS setup (25): (A) compressor, (B) die, (C) micro-syringe pump, (D) hypodermic syringe, (E) teflon tubing, (F) blunt needle, (G) cylindrical frame, (H) power supply, (I) receiver, (J) cross section of the die.

because of the repulsion of the like charges (14,25). CSBS nanofibers are formed under the combination of airflow stretching force and electric field force (25).

2.3 Experimental details

Two groups of experiments were carried out according to the process parameters listed in Tables 1 and 2. The fabrication time for each experiment is 80 min. The prepared samples were sputtered with sputter coater (SBC-12, China Science Instruments Co., Ltd., China), and then the morphology of the samples was photographed with scanning electron microscopy (SEM; S-3400, Hitachi; High-Technologies, Japan). Four SEM photos were taken for each sample. At least 100 measure points were randomly selected for each sample; the fiber diameter at each measure point was measured using ImageJ (National Institutes of Health, USA). The average diameter and diameter standard deviation of each sample were calculated. Ghasemi-Mobarakeh *et al.* method (32) was used to measure the porosity of each sample.

Table 1: Spinning conditions of different air pressures

Parameter	Values
Voltage (kV)	7
Length of cylinder (cm)	10
Diameter of cylinder (cm)	15
Needle to cylinder distance (cm)	8
Left face of cylinder to collector distance (cm)	95
Injection speed (mL/h)	0.5
Air pressure (MPa)	0.007, 0.01, 0.013, 0.016, 0.019

Table 2: Spinning conditions of different injection speeds

Parameter	Values
Voltage (kV)	10
Length of cylinder (cm)	10
Diameter of cylinder (cm)	15
Needle to cylinder distance (cm)	6
Left face of cylinder to collector distance (cm)	95
Injection speed (mL/h)	0.2, 0.3, 0.4, 0.5, 0.6
Air pressure (MPa)	0.01

2.4 Numerical simulation

Part (j) in Figure 1 depicts a cross section of the CSBS die used in this article. The left side is the high-pressure gas inlet, and the right side is the gas outlet. Figure 2 shows the boundary conditions and the computational domain. The computational domain was drawn based on the size of the die. OG (= 2 mm) was the pressure inlet; GF (= 3 mm), EF (= 3 mm), ED (= 80 mm), and CD (= 40 mm) were the wall; CB (= 120 mm) and BA (= 105 mm) were the pressure outlet; OA was the symmetry; $\angle BCD = 120^\circ$. It can be seen from part (j) in Figure 1 that the cross section

Figure 2: Boundary conditions and computational domain.

of the die is symmetrical. To save calculation time, half of the airflow field was simulated.

The model was generated in SpaceClaim 2020R2, and the airflow field was simulated by FLUENT 2020R2. The k - ϵ model was used in this paper (18,33). The five air pressure values (0.007, 0.01, 0.013, 0.016, and 0.019 MPa) in Table 1 were selected as the inlet air pressures. The outlet air pressure was set to normal pressure. The number of iterations was set to 1,000. The residuals of the five model equations of continuity, x -velocity, y -velocity, k , and epsilon were all set to 10^{-4} , and the residuals of the energy equation was set to 10^{-6} .

3 Results and analysis

3.1 The effect of air pressure on the fabrication of CSBS fibers

As shown in Figure 3, an increase in air pressure will cause a smaller fiber diameter. This conclusion can also be drawn from Figure 4: When the air pressure increased from 0.007 to 0.019 MPa, the average diameter of CSBS fibers decreased from 1,662 to 990 nm. In addition, Figure 4 shows that as the air pressure increased from 0.007 to 0.019 MPa, the porosity of the nonwovens decreased from 45.85% to 36%.

Different from traditional SBS and ES, the CSBS fabricates nanofibers with simultaneous air stretching and electrostatic force. The following are the explanations of the above experimental results from the effect of airflow field and electric field on jet.

3.1.1 Effect of airflow field on fiber preparation under different air pressures

Figure 5 shows the contours of velocity field under different air pressures. The parts selected with oval windows in Figure 5 are the partial enlarged views of the inlet of each velocity fields. Figure 5 depicts that an increase in air pressure will cause an increase in air velocity. As the tip of the blunt needle in the CSBS setup is always facing the center of the die's outlet, the jet fly roughly along OA (Figure 2). In addition, many articles have studied the physical quantities on the centerline of the airflow field to reveal the mechanisms of polymer fiber preparation (18,33–36). To investigate the impact of CSBS airflow on jets and fibers, the centerline velocities of each model

Figure 3: SEM photos of CSBS fibers prepared at the air pressures: (a) 0.007 MPa, (b) 0.01 MPa, (c) 0.013 MPa, (d) 0.016 MPa, (e) 0.019 MPa.

Figure 4: The relationships between CSBS fiber diameter, CSBS nonwoven porosity, and air pressure.

were extracted and plotted as in Figure 6a. Figure 6a shows that the centerline velocity increases as the air pressure increases. High-speed gas can only fully stretch the jet within the effective stretching distance (100 μm from the tip of the needle) (24). When the jet leaves the effective stretching distance, the high-pressure gas hardly affects the stretching of the jet. Therefore, the centerline velocities within the effective stretching distance of the five models were extracted and plotted as in Figure 6b. The part selected with the red window in Figure 6b is the effective stretching distance of the airflow. Figure 6b

shows that the greater the air pressure, the greater the centerline air velocity within the effective stretching distance. In addition, Chung and Abdalla (37) found the following relationship between the air force exerted on the polymer jet (or fiber) and the air velocity:

$$dF = \frac{c\rho\pi^{0.815}\mu^{0.61}Q^{0.815}(U - V_L)^{1.39}dx}{V_L^{0.815}}, \quad (1)$$

where F is the airflow drag force, C is a constant, x is the fiber/jet axis, μ is the air kinetic viscosity, ρ is the air density, U is the air velocity, Q is the polymer volume flow rate, and V_L is the final fiber velocity.

From Eq. 1, the faster the air velocity (U), the stronger the stretching force (F) of the airflow on the jet. In combination with the conclusion in Figure 6b, it can be seen that the higher the air pressure, the faster the airflow speed, and the greater the air force exerted on the CSBS jet within the effective stretching distance. Thus, increasing the air pressure can increase the stretching force of the airflow on the jet to produce thinner CSBS fibers.

Extract the centerline turbulence intensities of the five models and plot these data as in Figure 7a. The turbulence intensities in the effective stretching distance in Figure 7a were extracted again and plotted as in Figure 7b. The part selected with the red window in Figure 7b was the effective stretching distance of the high-speed gas. Figure 7b shows that the greater the air pressure, the greater the centerline turbulence intensity within the

Figure 5: Contours of velocity field under different air pressures: (a) 0.007 MPa, (b) 0.01 MPa, (c) 0.013 MPa, (d) 0.016 MPa, (e) 0.019 MPa.

Figure 6: Centerline velocities under different air pressures: (a) research distance is $[0, 0.187 \text{ m}]$, (b) research distance is the effective stretch distance.

effective stretching distance. The smaller turbulence velocity fluctuations are beneficial to prevent the instability in fiber formation (33). Therefore, the greater the air pressure, the greater the intensity of the turbulence, which is more likely to cause the instability in fiber formation.

Gholipourmalekabadi et al. found that the thinner the fiber, the lower the porosity of the fiber web (38). Consequently, the increase in air pressure causes the CSBS fibers to become thinner, which leads to a decrease in the porosity of the nonwoven.

3.1.2 Effect of electric field on fiber preparation under different air pressures

In CSBS system, when the jet enters the electric field formed by the cylindrical electrode, it will be charged with the same kind of charges because of electrostatic induction. These like charges will generate charge repulsion within the jet. When the charge repulsion exceeds the surface tension of the jet, the jet will split and become thinner (25). In our previous research, the formula for the surface charge density of the CSBS jet was obtained:

Figure 7: Centerline turbulence intensities under different air pressures: (a) research distance is $[0, 0.187 \text{ m}]$, (b) research distance is the effective stretch distance.

$$\sigma_{\text{jet}} = \frac{Q_{\text{jet}}}{S_{\text{jet}}} = \frac{\varepsilon_0 U_0}{r_1 \ln \frac{r_2}{r_1}}, \quad (2)$$

where σ_{jet} is the surface charge density of jet; ε_0 is the permittivity of free space; U_0 is the voltage; r_1 is the radius of the jet; and r_2 is the radius of the cylindrical electrode (39). The permittivity of free space (ε_0) is a constant. From the parameters in Tables 1 and 2, it can be seen that for a group of experiments in this article, the voltage (U_0) and the radius of the cylindrical electrode (r_2) remain unchanged. Therefore, in Eq. 2, the jet radius (r_1) is the only independent variable and the jet surface charge density (σ_{jet}) is the only dependent variable.

Denote the denominator of Eq. 2 by $x(r_1)$, then:

$$x(r_1) = r_1 \ln \frac{r_2}{r_1}. \quad (3)$$

Take the derivate of Eq. 3 with respect to r_1 :

$$x'(r_1) = \ln \frac{r_2}{r_1} - 1. \quad (4)$$

The unit of the radius of the cylindrical electrode (r_2) is centimeter, as shown in Tables 1 and 2. Bolbasov et al. found that within the effective stretching distance, the airflow can stretch the diameter of the jet to less than 1/100 of the inner diameter of the needle (24). The inner

Figure 8: SEM photos of fibers prepared at the injection speeds: (a) 0.2 mL/h, (b) 0.3 mL/h, (c) 0.4 mL/h, (d) 0.5 mL/h, (e) 0.6 mL/h.

diameter of the needle used in this work was 0.42 mm. Therefore, when the jet entered the cavity of electrode, the radius of the jet was in the micron scale. Thus, $\frac{r_2}{r_1} \gg e$ ($= 2.71828\dots$) and $\ln \frac{r_2}{r_1} \gg 1$. Therefore, $x'(r_1)$ is a positive number and Eq. 3 is a monotonous increasing function. The decrease in jet radius (r_1) will lead to the decrease in $x(r_1)$ and the increase in σ_{jet} . The larger the σ_{jet} is, the more likely the jet is to split and thin because of charge repulsion. Consequently, the smaller the jet diameter, the greater the surface charge density of the jet, which makes the jet in the CSBS electric field easier to split because of the repulsive force among the charges. Increasing the air pressure can make the jet diameter smaller. Thus, the increase in air pressure not only facilitates the stretching and thinning of the jet by the air flow, but also helps the jet to become thinner because of charge repulsion. In summary, increasing the air pressure can reduce the CSBS fiber diameter.

3.2 The effect of injection speed on the fabrication of CSBS fiber

Figure 8 depicts the morphologies of CSBS fibers prepared according to Table 2. Figure 9 shows that as the injection speed decreases from 0.6 to 0.2 mL/h, the average fiber diameter decreases from 1,429 to 781 nm, and the porosity of the nonwoven decreases first and then increases. The reasons for these results are as follows.

Figure 9: The relationships between CSBS fiber diameter, nonwoven porosity, and injection speed.

The reduction in the injection speed will cause the initial volume of the jet to become smaller and thus to form a thinner jet. Furthermore, Eq. 2 and its analysis results showed that the thinner the jet is, the more likely it is to generate higher surface charge density and thus to prepare thinner fibers. Therefore, reducing the injection speed can not only reduce the initial jet diameter, but also facilitate the thinning of the jet in the CSBS electric field, thereby preparing thinner nanofibers.

For the five experiments in Figure 8, there are two main factors affecting the porosity of CSBS nonwovens: fiber diameter and fiber production. The smaller the fiber diameter, the smaller the porosity of the nonwoven (38). Too little fiber production will result in greater porosity. This is because too few fibers cannot take up more space. The preparation time of the samples in this paper is the same; therefore, the lower the injection speed, the less the fiber production. The result of the competition between the above two factors determines the porosity of the nonwoven. Therefore, the porosity curve in Figure 9 showed a decreasing trend first and then increasing with the increase in injection speed.

4 Conclusion

This paper used computational fluid dynamics technique to study the CSBS airflow field. The simulation results showed that as the air pressure increases, the centerline velocity and the centerline turbulence intensity within the effective stretching distance increase. The increase in the centerline velocity will increase the stretching force of the air-flow on the jet, which will make the jet thinner. The increase in centerline turbulence intensity will increase the instability in the formation of CSBS fiber. The theoretical study of the CSBS electric field revealed that the surface charge density of the CSBS jet increases with the decrease in the jet diameter. The increase in air pressure can not only increase the stretching of the jet by the air flow, but also make the jet thinner because of the increase in surface charge density. Increasing the air pressure can reduce the CSBS fiber diameter and the porosity of the nonwoven. The reduction in injection speed can reduce the initial diameter of the jet and increase the surface charge density of the jet. Therefore, reducing the injection speed can make the diameter of the CSBS fiber smaller. Under the competition of fiber diameter and fiber production, the porosity of the nonwoven decreases first and then increases with the increase in the injection speed.

In this work, a combination of numerical simulation, theoretical analysis, and experiment is used to study the effects of CSBS airflow field and electric field on the fabrication of CSBS nanofibers for the first time. The conclusions of this article provide theoretical and experimental bases for the controllable preparation of CSBS nanofibers, which are helpful for the application and promotion of CSBS technology in battery, filtration, membrane, etc.

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