*Journal of
Nanoscience and Nanotechnology*
Vol. 7, 2667–2673, 2007

Gas Flow-Assisted Alignment of Super Long Electrospun Nanofibers

Weiping Zhou, Zhifei Li, Qiang Zhang, Yanping Liu, Fei Wei*, and Guohua Luo

*Beijing Key Laboratory of Green Reaction Engineering and Technology, Department of Chemical Engineering,
Tsinghua University, Beijing 100084, P. R. China*

Electrospinning provides a simple and versatile method for generating ultra thin fibers with diameters ranging from nanometer to micron out of various materials. However, there are still challenges in the alignment of electrospun nanofibers, which is an important step toward the exploitation of these fibers in applications. In this letter, we report a method using the gas flow to assist the alignment of electrospun nanofibers, which can form well-aligned super long polymeric nanofibers over large areas with the length of more than 20 cm. The improved collector is built by coupling a "T"-shaped electrode and a rectangle electrode, and it can make the electrospun nanofiber form a fixed site at the "T"-shaped electrode under the electric field and make it possible to use an assisting gas flow (AGF) to draw the other part of the nanofiber to fly toward the upside of the rectangle electrode and obtain well-aligned long nanofibers. These well-aligned long nanofibers can be further applied easily without disturbing the aligned structure, which is convenient for the measurement and device fabrications.

Keywords: Electrospinning, Alignment, Assisting Gas Flow.

1. INTRODUCTION

In recent years, electrospinning has become more and more popular due to providing a straightforward way of producing polymer fibers with diameters as small as several nanometers and up to a few microns. These ultra thin fibers have potential applications in various areas such as chemical sensors, electronic devices, filters, nanocomposites, smart textiles, etc.[1–3] In the process of electrospinning, a solid fiber is generated as the electrified jet (composed of a suitable viscous polymeric solution) is continuously stretched due to the electrostatic repulsions between the surface charges and the evaporation of solvent.[4,5] As limited by the chaotic motion (or bending instability) of the highly charged jet, the electrospun fibers are often collected as randomly oriented structures in the form of nonwoven mats. Although these mats are of great interests for applications including texturing, composite reinforcement, membrane-based separation and tissue engineering,[6–8] their disordered structures seem to be limited for the application in device fabrications that often require well-aligned and highly ordered architectures, such as microelectronics and photonics.[9,10]

Up to now, some great progress has been made in the control of the spatial orientation of electrospun fibers. Aligning these fibers during electrospinning process is difficult because the method involves the application of very high electric voltages (10–30 kV) and big distances (15–30 cm) between the electrodes. Researchers have explored novel methods including both mechanical and electrostatic ways to align polymeric and inorganic nanofibers. Sundaray et al. have shown that as-spun fibers could be aligned (parallel and cross patterns) when an insulated cylinder rotating with a high speed was used as the collector.[11] With the similar method, Katta et al. used copper wires spaced evenly in the form of a circular drum as a collector of the nanofibers. However, the degree of alignment of the nanofibers was seriously influenced by the collecting time. With the increment of the collecting time those nanofibers would become more and more disordered.[12] Theron et al. fabricated an electrostatic field-assisted assembly technique combined with an electrospinning process used to position and align individual nanofibers on a tapered and grounded wheel-like bobbin. But they could only attain aligned nanofibers in very small regions and the collector was not easy to prepare.[13] Li et al. have prepared polymeric and ceramic nanofibers as axially aligned arrays by using of a collector with two pieces of electrically conductive substrate separated by a gap. The electrospun fibers would tend to align across the electrodes, thus creating a pattern. But this method also has some deficiencies, such as the distance of the gap could

---

*Author to whom correspondence should be addressed.

not be too big and the degree of the alignment would be evidently affected by the collecting time as well.[14] Smit et al. designed a technique to make continuous uniaxial fiber bundle yarns from electrospun, which consisted of spinning onto a water reservoir collector and drawing the resulting non-woven web of fibers across the water before collecting the resulting yarn.[15] Teo et al. described a novel counter-collector made of two steel blades which were placed in line with a gap between them and could collect a fiber bundle of micron size diameter with aligned nanofibers within the gap.[16] And most recently, Chuangchote et al. reported a new technique called dual vertical wire technique. The application of the technique on polyvinyl alcohol (PVA) to prepare uniaxially-aligned fibers was found to be successful at short collection times. However, further increase in the collection time resulted in some of these aligned fibers binding together to form a large nanofibrous fiber tow.[17] As most of these collectors can only obtain fiber bundles or fiber tow, there are many limitations in the further applications.

Though there has been significant progress in alignment of electrospun nanofibers, most of the collectors are relatively complicated and the lengths of those nanofibers are almost all shorter than one centimeter except those fiber bundles. Here we report a simple and versatile method that can generate well-aligned nanofibers over large areas with the length of more than 20 cm by introducing creative collectors and using of a gas flow to assist the alignment of nanofibers. In this work, we have designed two sets of simple electrospinning setups, both of which are very easy to be prepared and can attain aligned nanofibers. The collecting electrode of setup 1 is made up of two or three pieces of coaxial circle metal sheets, which can easily collect aligned nanofibers. These metal sheets together with the aligned nanofibers on it can construct a two-floor or three-floor tapered tower. However, because the degree of alignment of these nanofibers collected by setup 1 are not perfect, setup 2 has been designed based on setup 1 by introducing a "T"-shaped electrode and a rectangle electrode. Furthermore, a gas flow is added to assist the alignment of the electrospinning nanofibers in setup 2. The gas flow or air-blowing has already been used and researched in electrospinning process,[18–20] and the experimental results demonstrated that it could provide many advantages, such as controlling the stability of Taylor cones, accelerating the solvent evaporation process, decreasing the solution viscosity at the spinneret and changing the fiber diameter by controlling the air temperature, the direction of air-flow and the air flow rate. In this work, the gas flow is used to control the direction of nanofibers' motion and get well-aligned nanofibers. During the electrospinning process, the electrospun nanofiber can form a fixed site at the "T"-shaped electrode under the action of electric field and the AGF helps the other side of the nanofiber to fly toward the upside of the rectangle electrode. Thus, well-aligned nanofibers over large areas with super long lengths can be obtained on the collecting electrode of setup 2. Further, by adjusting the size of the collector of setup 2, different lengths of well-aligned electrospun nanofibers can be obtained easily. These well-aligned super long nanofibers are convenient for further applications, such as chemical sensors, electronic devices, and for the measurement of their mechanical and electrical properties without disturbing the aligned structure.

2. EXPERIMENTAL DETAILS

Figures 1 and 2 present the schematic illustration of the two setups for electrospinning used in this letter. For both of setups, the positive electrode connected to the spinneret is a circle metal sheet with the diameter of 10 cm. In the setup 1, the grounded electrode for collecting nanofibers consists of two or three pieces of coaxial metal sheets, the diameters of which are 1 cm, 1.5 cm, and 10 cm, respectively, and the distance between each two neighboring pieces is about 1 cm. In order to use the gas flow to assist the alignment of nanofibers, the collecting electrode of setup 2 is made up of a "T"-shaped electrode and a rectangle electrode. The direction of gas flow is along the inclined plane determined by the "T"-shaped electrode and the upside of the rectangle electrode. The electrospun nanofibers will be up-displaced because of the AGF's influence, so the position of the collector is several centimeters higher than that of the positive electrode in setup 2, as shown in Figure 2.

The polymeric solution used for electrospinning is polyethylene oxide (PEO) ($M_w = 900,000$ g/mol) aqueous solution with weight concentration of 3–3.5%. Electrospinning was carried out by using air pressure of 0.1–0.3 kg/cm^2 to force the solution to spray out of a spinneret with the diameter of 1 mm at a voltage of 30 kV. The distance between the positive electrode and the grounded collector was 25 cm. During the whole electrospinning process the ambient temperature was kept at about 40 °C.

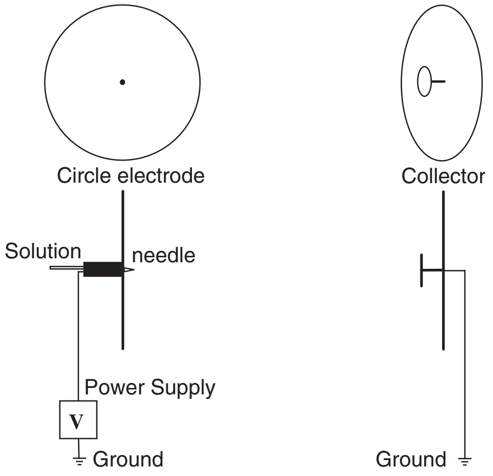

Fig. 1. The sketch of setup 1 for electrospinning.

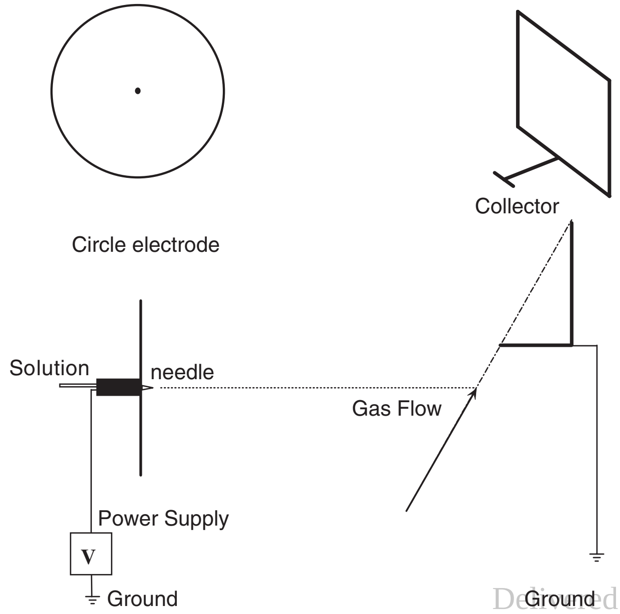

Fig. 2. The sketch of setup 2 for electrospinning.

The nanofibers on collectors could be easily put onto the silicon wafers or copper grids coated by the holy carbon film. Then the samples were directly used for the characterization by Scanning Electron Microscopy (SEM) (JSM 7401F) and High-Resolution Transmission Electron Microscopy (HRTEM) (JEOL 2010, exciting at 200 kV) without any morphological changes.

3. RESULTS AND DISCUSSION

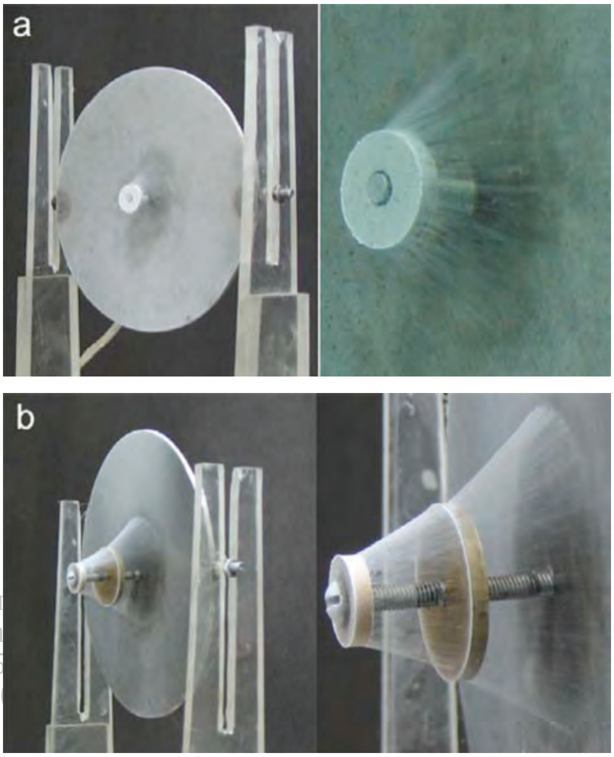

Fig. 3. The collector of setup 1 with aligned nanofibers: (a) the collector with two pieces of circle metal sheets; (b) the collector with three pieces of circle metal sheets.

The nanofibers collected by setup 1 are shown in Figure 3, which formed a very regular shape like a two-floor or three-floor tapered tower on the circle collector. All the nanofibers radially extended from the smaller piece of metal sheet to the larger ones. Figure 4 shows the cross-sectional view of the electric field strength vectors between the positive electrode and grounded collector calculated by the Student's QuickField program. Unlike the conventional system, the electric field lines in the vicinity of the collector are concentrated towards to the smaller circle metal sheet. As shown in Figure 5, when the nanofiber with positive charges is flying to the grounded collector by the electrical driving force, it will prefer to move along the electric field lines, flying toward the smaller circle metal sheet. After the front point of the nanofiber has touched the smaller circle metal sheet, the positive charges on this part will be counteracted immediately by the negative charges on the grounded collector. Because the polymer nanofiber is insulated, the other part of the nanofiber far from the collector still has some residual positive charges, and it will keep on moving under electrical driving force until it reaches the larger circle metal sheet. According to the principle above, it is easy for the nanofibers to extend from smaller circle metal sheet to bigger ones and realize the alignment along the radial direction of the circle metal sheets. Observed from SEM (Fig. 6), it was found that there were some irregularly aligned nanofibers though most nanofibers oriented along the radial direction of circle collectors. This was because after the front part of the nanofiber had touched the smaller circle metal sheet, the positive charges on the other part would gradually fade during this part flying toward the larger circle metal sheet, and the electrical driving force would also gradually decrease. Additionally, the nanofiber was not rigid and its movement would be influenced by many factors.

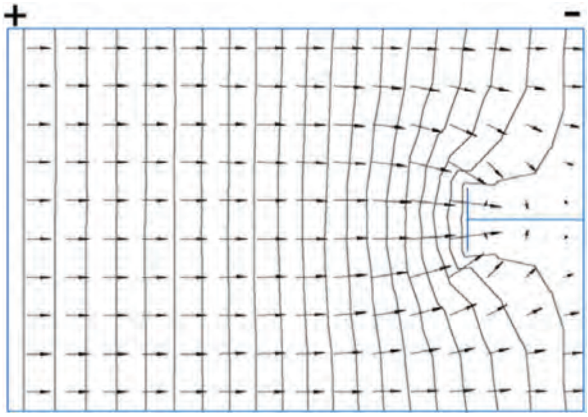

Fig. 4. Calculated electric field strength vectors in the region between the positive electrode and the grounded collector.

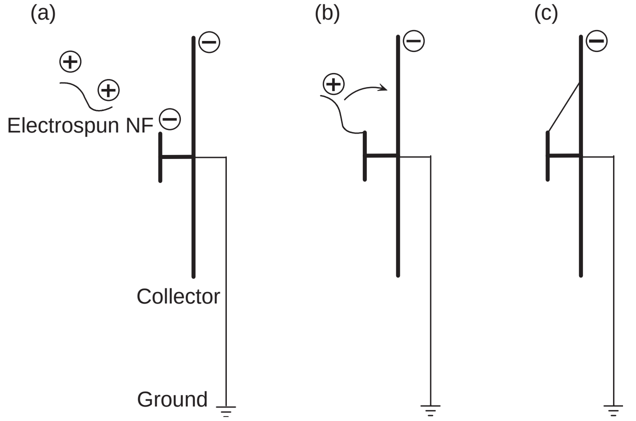

Fig. 5. The sketch of the orientation of the electrospun nanofiber on the collector of setup 1: (a) an electrospun nanofiber with positive charger is flying to the collector; (b) the front point of the electrospun nanofiber touches the smaller circle metal sheet and becomes a fixed site; (c) the end of the electrospun nanofiber flies to the bigger circle metal sheet.

Therefore, it is almost impossible to obtain well-aligned nanofibers only by electric field.

In order to improve the degree of alignment of nanofibers, setup 2 was designed on the base of setup 1, which is made up of a protrudent "T"-shaped electrode and a rectangle electrode. The experimental result showed that almost all the nanofibers were randomly collected on the "T"-shaped electrode and couldn't reach the rectangle electrode without any other assistant method. So an assisting gas flow (AGF) is used to draw the other part of the nanofiber to fly toward the upside of the rectangle electrode. As shown in Figure 2, the direction of the AGF is parallel to the inclined plane determined by "T"-shaped electrode and the upside of the rectangle electrode. With the similar principle to setup 1, when the electrospun nanofiber with positive charges is flying toward the collector (Fig. 7(a)), the front point of the nanofiber will touch the protrudent "T"-shaped electrode first under the co-action of the electric field and the AGF (Fig. 7(b)), then the other part of the nanofiber keeps on moving to the upside of the rectangle electrode along the direction of the AGF (Fig. 7(c)) until the highest part of the nanofiber reaches the upside of the rectangle (Fig. 7(d)). At the same time, a new point on the nanofiber comes to the "T"-shaped electrode and forms another fixed point on the nanofiber, then the same scene in Figure 7(c) occurs again. In addition, the polymer electrospun nanofiber is insulated, so the remaining positive charges on the nanofibers will not immediately disappear and the electrostatic repulsion between the deposited and the upcoming fibers can further enhance the parallel alignment. Thus, well-aligned electrospun nanofibers can be obtained on the collector. As demonstrated in Figure 8 (the side lengths of the rectangular frame are 15 and 8 cm and the dimension of the "T" shape is 6 and 3 cm), it was very easy to obtain well-aligned electrospun nanofibers over very large areas with the length over 10 cm. By changing the size of the collector (the side lengths of the rectangular frame are 19 and 15 cm and the dimension of the "T" shape is 8.5 and 3 cm) and adjusting the speed of the AGF, the well-aligned nanofibers with the length of over 20 cm can also be attained (Fig. 9).

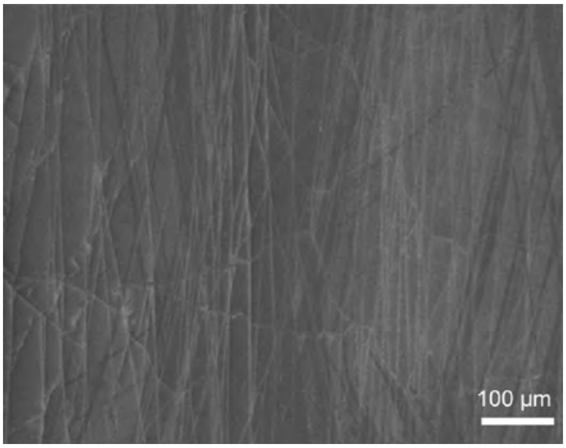

Fig. 6. The SEM image of aligned nanofibers collected by setup 1.

Observed from SEM, it was found that these well-aligned nanofibers in macro scale also showed the perfect alignment in micro scale (Figs. 10(a)–(b)). And according to TEM images, such as Figure 11, the statistical result showed the average diameter of the PEO nanofibers was about 80 nm. In order to study the influence of collecting time, electrospun samples collected by setup 2 were analyzed at various time increments in the electrospinning process ranging from 10 sec to 2 min. As seen in the SEM images, good alignment is present up to approximately 1 min (Figs. 10(a)–(e)). After 2 min, the mat of nanofibers became thick and entanglements form (Fig. 10(f)). This experimental result about the collecting time is similar with that of Katta et al.[12] In this work, there are two possible reasons for the loss in alignment with thicker mats. Firstly, with the increment of time, more and more non-conductive nanofibers deposited on the "T"-shaped electrode, which could obstruct the counteraction of positive charges on nanofibers and negative charges on the electrode, and result into an accumulation of positive charges in the nanofiber mat. So the attraction between the "T"-shaped electrode and the coming nanofibers became weaker and it was harder for the front end of the nanofiber to touch the protrudent "T"-shaped electrode. The second reason possibly resulted from the disturbance of gas flow, which also could affect the alignment of the nanofibers. Thus, for setup 2 in this work, better aligned electrospun nanofibers can be obtained by controlling the collecting time less than 2 min and improving the stability of the gas flow.

During the electrospinning process of using setup 2, the AGF, besides the design of the collector, is the most important to obtain well-aligned electrospun nanofibers. Firstly, the direction of the AGF must be parallel to the inclined

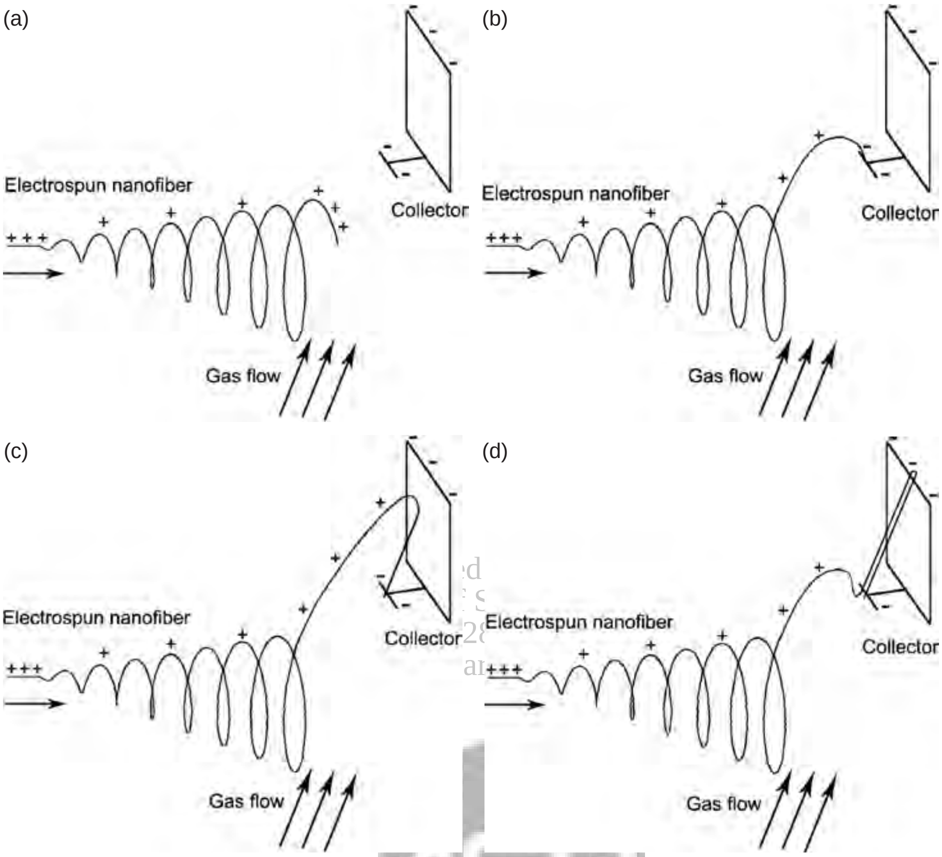

Fig. 7. The sketch of the orientation of electrospun nanofiber on the collector of setup 2.

plane determined by "T"-shaped electrode and the upside of the rectangle electrode, which is necessary to lead the other part of the nanofiber to fly toward the upside of the rectangle electrode after one end of the nanofiber has become a fixed site at the "T"-shaped electrode. Secondly, the speed of the AGF must be suited with the polymeric solution and the distance between the "T"-shaped electrode and the upside of the rectangle electrode. On one hand, if the speed of the AGF is too low, it can not lead the highest point of the electrospun nanofiber to reach the upside of the rectangle electrode. On the other hand, if the speed of the AGF is too high, it will result in the broken of the electrospun nanofiber and blow the nanofiber much far from the upside of the rectangle electrode. Thus, it is significant to find out an optimized speed of the AGF for different polymeric systems and collectors by experiments. For our polymeric system, polyethylene oxide (PEO) aqueous solution with its weight concentration of 3–3.5%, the suitable speed of AGF at the "T"-shaped electrode is about 4 m/s for the collector in Figure 8 and 6 m/s for the collector in Figure 9, respectively. For both collectors (in Figs. 8 and 9) the nanofibers are easily broken when the speed of the gas flow is beyond 12 m/s.

In addition to the materials described in this paper, we have also successfully obtained well-aligned nanofibers with some other kinds of polymers, such as polyvinyl alcohol (PVA) (M_w = 75,000–79,000, hydrolysis 98–99%) with the weight concentration of about 4–6%.

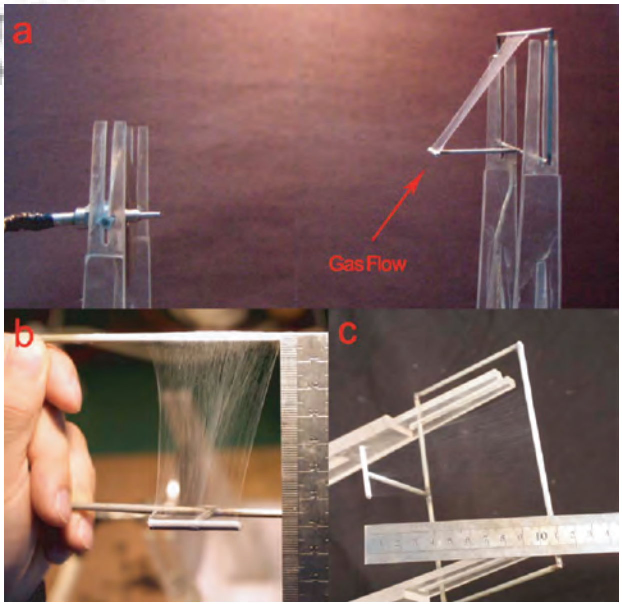

Fig. 8. (a) setup 2 for electrospinning; (b, c) the collector with the length of nanofibers over 10 cm.

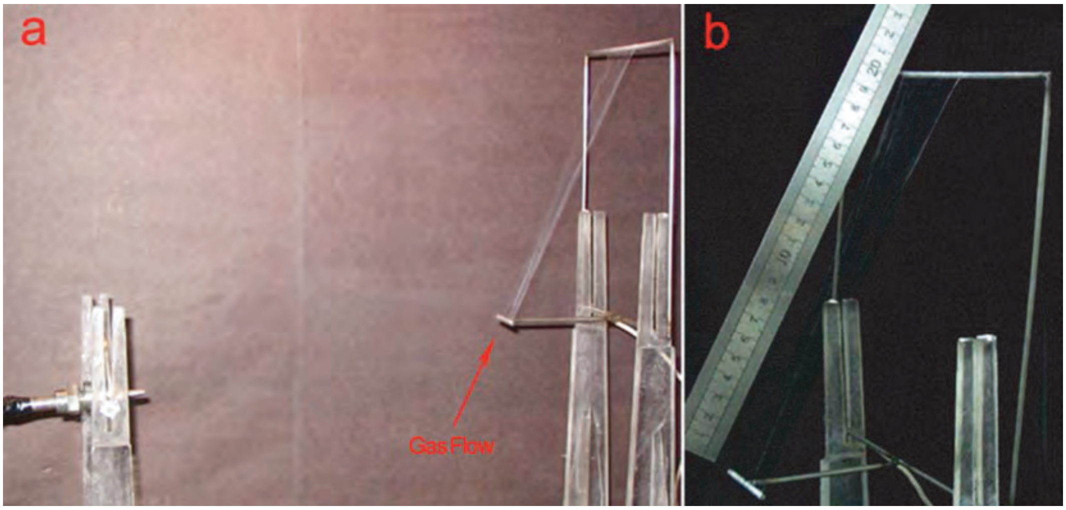

Fig. 9. (a) setup 2 for electrospinning; (b) the collector with the length of nanofibers over 20 cm.

Furthermore, the collector of setup 2 can be changed to many other forms if only following the same principle and using a suitable gas flow to control the collecting direction. For example, Figure 12 gives another design of setup 2.

These well-aligned long nanofibers collected by setup 2 are very convenient for further measurements of electric, optical, and mechanical properties without disturbing the aligned structure. And they also have potential applications

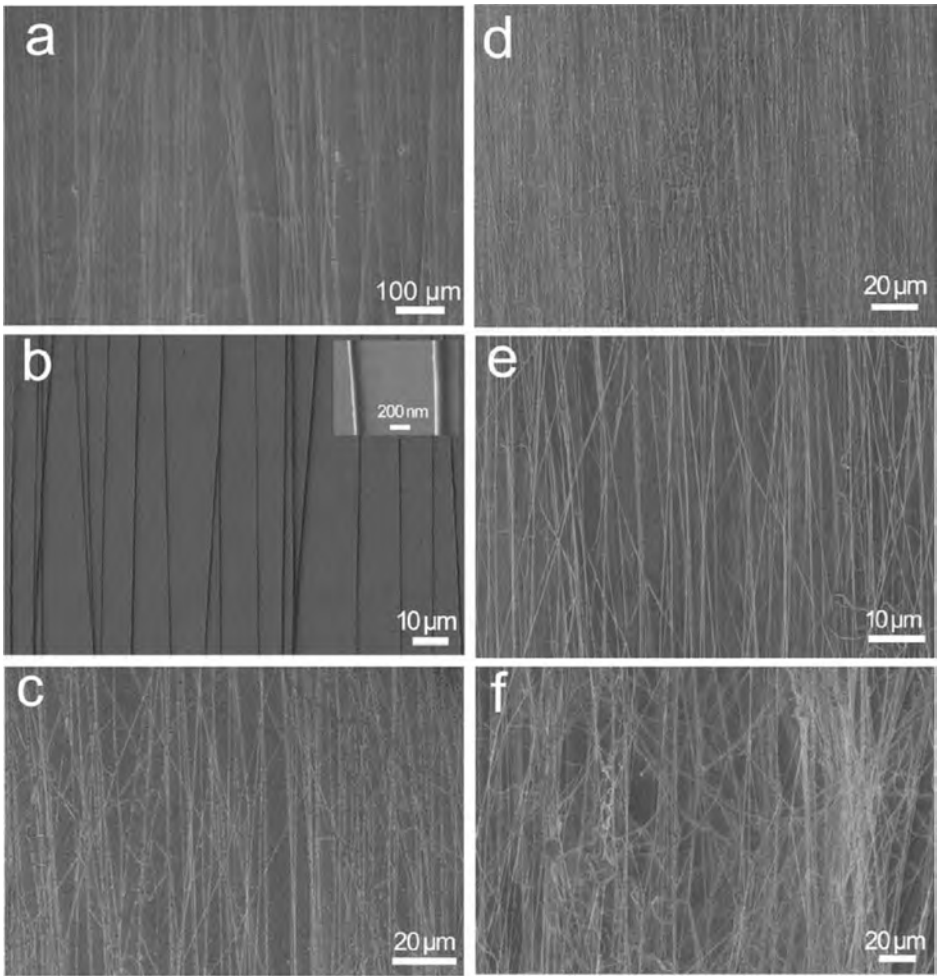

Fig. 10. The SEM images of well-aligned nanofibers collected by setup 2: (a, b) alignment after 10 s of spinning time (the inset in (b) shows enlarged SEM image of the nanofibers.); (c) after 30 s; (d, e) after 1 min; (f) after 2 min.

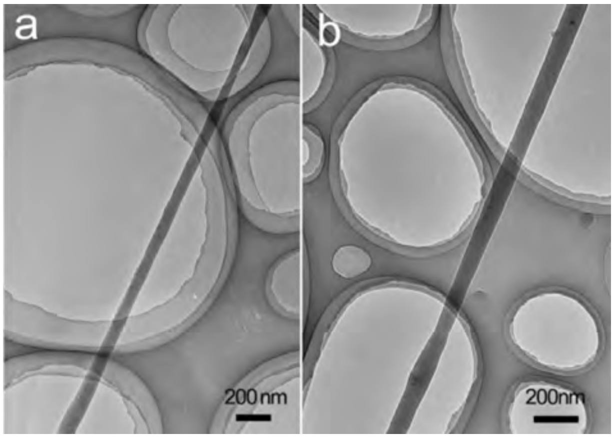

Fig. 11. The TEM images of single PEO nanofibers.

Fig. 12. The changed collector of setup 2.

in composite materials, reinforcements, electrochemical sensing, and tissue engineering, which often require well-aligned and highly ordered architectures.

4. CONCLUSION

In this work, we successfully designed two electrospinning setups to collect aligned nanofibers. The experimental results have demonstrated the effectiveness of these two setups for assembling nanofibers in parallel arrays. Especially for setup 2, with the gas flow to assist the alignment, it can easily control the collecting direction of nanofibers and obtain well-aligned nanofibers in very macro scale. We believe this simple and versatile electrospinning setup with a convenient control approach of a gas flow will bring many advantages to the electrospinning process and create more novel applications for the nanofibers.

Acknowledgments: This work is supported by the National High Technology Research and Development Program of China (863 program, 2003AA302630) and the Natural Science Foundation of China (20236020).

References and Notes

1. X. Wang, C. Drew, L. Soo-Hyoung, K. J. Senecal, S. Tripathy, and L. Samuelson, *Nano Lett.* 2, 1273 **(2002)**.
2. M. M. Burgshoef and G. J. Vancso, *Adv. Mater.* 16, 1362 **(1999)**.
3. S. Aryal, N. Dharmaraj, S. R. Bhattarai, M. S. Khil, and H. Y. Kim, *J. Nanosci. Nanotechnol.* 6, 510 **(2006)**.
4. W. M. Edwards, U. S. Patent No. 3179614 **(1965)**.
5. R. S Irwin, U. S. Patent No. 3415782 **(1968)**.
6. K. J. Pawlowski, H. L. Belvin, D. L. Raney, J. Su, J. S. Harrison, and E. L. Siochi, *Polymer* 44, 1309 **(2003)**.
7. G. E. Wnek, M. E. Carr, D. G. Simpson, and G. L. Bowlin, *Nano Lett.* 3, 213 **(2003)**.
8. P. Wutticharoenmongkol, N. Sanchavanakit, P. Pavasant, and P. Supaphol, *J. Nanosci. Nanotechnol.* 6, 514 **(2006)**.
9. Y. Huang, X. Duan, Q. Wei, and C. M. Lieber, *Science* 291, 630 **(2001)**.
10. F. Favier, E. C. Walter, M. P. Zach, T. Benter, and R. M. Penner, *Science* 293, 2227 **(2001)**.
11. B. Sundaray, V. Subramanian, T. S. Natarajan, R. Z. Xiang, C. C. Chang, and W. S. Fann, *Appl. Phys. Lett.* 84, 1222 **(2004)**.
12. P. Katta, M. Alessandro, R. D. Ramsier, and G. G. Chase, *Nano Lett.* 4, 2215 **(2004)**.
13. A. Theron, E. Zussman, and A. L. Yarin, *Nanotechnology* 12, 390 **(2001)**.
14. D. Li, Y. L. Wang, and Y. N. Xia, *Nano Lett.* 3, 1167 **(2003)**.
15. E. Smit, U. Buttner, and R. D. Sanderson, *Polymer* 46, 2419 **(2005)**.
16. W. E. Teo1 and S. Ramakrishna, *Nanotechnology* 16, 1878 **(2005)**.
17. S. Chuangchote and P. Supaphol, *J. Nanosci. Nanotechnol.* 6, 125 **(2006)**.
18. I. C. Um, D. Fang, B. S. Hsiao, A. Okamoto, and B. Chu, *Biomacromolecules* 5, 1428 **(2004)**.
19. X. F. Wang, I. C. Um, D. F. Fang, A. Okamotoc, B. S. Hsiaoa, and B. Chua, *Polymer* 46, 4853 **(2005)**.
20. G. Larsen, R. Spretz, and R. Velarde-Oritz, *Adv. Mater.* 16, 166 **(2004)**.

Received: 29 May 2006. Accepted: 16 November 2006.