

Needleless Electrospinning: A Practical Way to Mass Production of Nanofibers

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Nanofibers produced by electrospinning have many unique characteristics, such as high surface-to-mass (or volume) ratio, ability to form a highly porous fibrous membrane with excellent pore interconnectivity, controllability in fiber diameter, surface morphology and fibrous structure, and easy to be functionalized by using functional polymers or adding functional chemicals into polymer solutions for electrospinning. These unique features have provided electrospun nanofibers with enormous opportunities to be used in various fields including tissue engineering scaffolds, filtration, catalyst and enzyme carriers, release control, sensors and energy storage, affinity membranes, recovery of metal ions, and many others [1-3].

The conventional approach to electrospinning nanofibers often uses a needle-like nozzle. Despite the enormous application potential, needle electrospun nanofibers meet difficulties in broad applications in practice, due to the lack of an economic and efficient way to scale up the electrospinning process.

Recently, needleless electrospinning has emerged as a new electrospinning mode and shown ability to produce nanofibers on largescales [4]. Needleless electrospinning is featured as electrospinning of nanofibers directly from an open liquid surface; numerous jets are formed simultaneously from the fiber generator without the influence of capillary effect that is normally associated with needle-like nozzles.

The first needleless electrospinning system was patented in 1979 [5], and it used a ring as spinneret for electrostatic production of fiber fleece. In 2005, a rotating roller was invented as fiber generator for mass electrospinning of nanofibers [6], and this technique was rapidly commercialized by Elmarco Co. with a brand name "Nanospider".

In needleless electrospinning, fiber generator plays a key role in determining the productivity and fiber morphology. According to the method to initiate the spinning process, needleless electrospinning can be classified into rotating or stationary electrospinning.

Rotating Needleless Electrospinning

Rotating needleless electrospinning uses mechanical vibrations to assist in the initiation of jets from polymer fluid surface. Several rotating spinnerets have been reported, including ring, ball, cylinder, coil and helical blade.

When the collector electrode is set on the uppermost of the rotary spinneret, nanofibers are generated upward and deposited on the collector. In this case, the spinneret is often immersed partially into a polymer solution underneath the spinneret. The rotation of spinneret has two functions: loading the polymer solution onto electrospinning sites and electrospinning the loaded solution into nanofibers. The later process is driven by a high electric field and enormous number of solution filaments is generated upward from the roller surface. Ring [5] and roller [6] setups belong to the category of upward rotary needleless electrospinning.

The jet formation in needleless electrospinning has been proposed to follow a few steps: 1) A thin layer of polymer solution is formed on the spinneret surface as a result of its partially immersion in the solution and rotation; 2) The rotation also causes perturbations on the solution layer thus inducing the formation of conical spikes on the solution surface; 3) When a high voltage is applied, the spikes concentrate electric forces thus intensifying the perturbations to form "Taylor cones"; 4) Jets are stretched out from the "Taylor cones" and finally result in fibers.

Niu et al. [7] compared rotating cylinder, ball and disc spinnerets using polyvinyl alcohol as model polymer. They also analyzed electric field in the electrospinning zone using a finite element method. Under the same working conditions, the disc produced finer nanofibers with a narrower diameter distribution compared to the ball and cylinder spinnerets. High electric field was found to be narrowly distributed on the disc top, which led to highly stretching of the solution jets. The cylinder spinneret had a large surface area, but the electric field distributed unevenly on the fiber-generating surface. The ball formed an electric field with low intensity thus generating fewer jets when compared to disc and cylinder generators. In comparison with the needle electrospinning, these spinnerets had much higher productivity.

Lin et al. [8] invented a new needleless electrospinning setup using a spiral coil as spinneret. They also compared the coil spinneret with rotary cylinder and disc spinners. The spiral coil had a higher fiber production rate than the cylinder spinneret of the same dimension [9]. The nanofibers produced from the coil spinneret were much thinner with a narrower fiber diameter distribution when compared with those from needle electrospinning. However, for disc, ball and cylinder spinnerets, they normally produced coarser nanofibers.

Rotating needleless electrospinning has been used to process different polymers, such as poly(vinyl alcohol) [7,10], polyacrylonitrile [11], polyurethane [12], carbon nanotube/poly(vinyl alcohol) [13], polyamic acid [14], polystyrene [15], cellulose acetate butyrate [16] and polypropylene [17]. Most of the processes were based on solution based electrospinning process except for polypropylene. It was reported that the solution concentration played a vital role in the electrospinning process [18]. When the polymer concentration was low, the chain entanglement was insufficient, which resulted in beads or beaded fibers instead of uniform fibers. However, if the polymer concentration was too high, the high viscosity restricted stretching of polymer

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fluid into fine filaments. In the range that a polymer solution could be successfully electrospun into uniform nanofibers, the variation of polymer concentration had a small effect on the fiber diameter [7].

In addition, needleless electrospinning can also be affected by other factors, including molecular weight of polymer, solution conductivity and surface tension, applied voltage, collecting distance and ambient environment (*e.g.* temperature, gas environment and humidity).

The flow rate in needleless electrospinning can't be controlled precisely, but could be adjusted in a very limited range through changing the rotating speed of spinneret. The rotating speed of a spinneret can be varied in large range from few rpm up to 200 rpm [7,11], depending on the spinning solution, spinneret dimension, and operating parameters. When the rotating speed was low, a thin solution layer was formed on spinneret surface. Electrospinning exhausted the solution layer until it reached a state of which the solution was too thin to be electrospun [19]. Increasing the rotating speed leads to a thicker solution layer, and solution processing can be thus increased. When the rotating speed is too high, the solution could be thrown out from the fiber generator surface. On the other hand, increasing the spinneret rotating speed also decreases the life span of "Taylor cones", resulting in a reduced productivity. In addition, the increase in the rotating speed of cylinder spinneret was reported to decrease the fiber diameter [11].

More recently, a horizontal bead chain was used to produce nanofibers in an upward fashion [20]. The chain comprises two parallel parts. The lower part was dipped in a polymer solution, while the higher part functioned to produce fibers. When a high voltage was applied, the beads on the top part of the chain generated fine fibers.

In addition, rotating spinnerets were also used for electrospinning nanofibers in a downward fashion. In this case, the solution was conveyed to the electrospinning sites through the action of both the spinneret rotation and the gravity. For example, Tang et al. [21] reported an electrospinning setup using a rotating roller as fiber generator. During electrospinning, the spinning solution was dropped to the roller. When the solution droplets attached to the electrically charged metal roller, jets were ejected and nanofibers were thus produced. This setup had a 24~45 times higher fiber productivity than a conventional needle electrospinning system.

Lu et al. [22] reported a needleless electrospinning system using a rotary cone as spinneret. Polyvinylpyrrolidone (PVP) solution was supplied to the cone by a glass pipe to ensure electrospin continuously. The PVP solution flowed down to the fiber generating sites (cone edge) along the cone surface under the gravity. Compared to the conventional needle system, this system produced fibers of a similar quality with 1000 times' larger production rate. When a high applied voltage was employed, more uniform nanofibers with smaller diameter were obtained.

When the polymer solution is conveyed under the assistance of gravity, the downward electrospinning performance becomes more dependent on the solution properties (e.g. viscoelasticity, surface tension) than that in the upward rotating electrospinning.

Stationary Needleless Electrospinning

Stationary needleless electrospinning is heavily relied on the initiation of jets from an open liquid surface. How to concentrate electric forces on the solution surface where jets are expected to

generate is the key to a successful electrospinning process. In stationary needleless electrospinning systems, conical spikes are often created with the aid of an external force, such as magnetic force, high pressure gas flow, and gravity.

In 2004, Yarin and Zussman [23] reported a needleless electrospinning system which used a magnetic field to initiate the jet formation. The setup comprised a bottom layer of ferromagnetic fluid and an upper layer of polymer solution. When an external magnetic field was applied to the fluid system and an electric field was added simultaneously to the polymer solution layer, the ferromagnetic fluid triggered the formation of steady vertical spikes, which perturbed at the interlayer interface. Under the action of a strong electric field, these spikes were drawn into fine solution jets. Compared with the multineedle electrospinning, the production rate of Poly Ethylene Oxide (PEO) nanofibers was 12 times higher. Nevertheless, electrospun nanofibers exhibited a relatively coarse fiber with large diameter distribution.

Blowing gas into a polymer solution was also used to initiate jet formation from a flat open solution surface. Such an electrospinning mode is also called "bubble electrospinning". During bubble electrospinning, when gas was introduced into a solution reservoir and at the same time a high voltage was applied, bubbles were generated on the solution surface. The gas bubbles made electric forces concentrate on the bubble surface, where initiated the formation of multiple jets [24].

Apart from electrospinning fibers upward, nanofibers can also be electrospun downward using some motionless setups. In these cases the gravity is utilized to assist in conveying the solution to the spinning sites. For example, Wang et al. [10] reported a conical coil spinneret which functioned as both solution container and fiber generator. When a high voltage was applied to the wire coil, solution was stretched out from both the wire surface and the gap between wires to form solution jets. This setup can improve the electrospinning productivity by 13 times and produce finer nanofibers compared with the conventional needle electrospinning.

In another setup reported by Thoppey et al. [25], a plate with a certain horizontal angle was used for retaining the spinning solution, and nanofibers were generated from the plate edge. An electrically insulated reservoir connected with one or more plastic pipettes was used to provide polymer solution to the plate, wherein each pipette formed a solution stream as a jet initiation site. The production rate of this electrospinning was over 5 times higher than that of the conventional needle electrospinning, even using a single spinning site (one pipette).

Wu et al. [19] reported an upward needleless electrospinning system using a stationary cylinder spinneret. PEO nanofibers produced by this setup were reported to be more than 260 times larger in production rate than that of the conventional needle electrospinning (0.02 g/h).

More recently, an aluminum bowl was used as a spinneret to electrospin nanofibers [26]. The jet initiation on the bowl surface was quite similar to that on the needle electrospinning. Nanofibers generated from the edge of the bowel were deposited on a concentric cylindrical collector. Nanofibers produced by this electrospinning system exhibited the same quality as those produced by conventional needle electrospinning, but a production rate was about 40 times higher.

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Conclusion

Needleless electrospinning shows great potential in electrospinning of nanofibers on large scales. Although diversely different spinnerets could be chosen for needleless electrospinning, they may not be ideal and some of them still need a further experimental verification in terms of the ability to control the fiber quality and electrospinning process. It still lacks extensive understanding on how the polymer types and solution properties, especially for those using organic solvent systems, affect the electrospinning process and productivity. It still remains challenges to electrospin bicomponent nanofibers using a needleless electrospinning technique. It is expected that with the further development of this electrospinning technology, high quality, low cost nanofibers will be widely used in our daily life in the not far future.

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