High-Throughput Needleless Electrospinning of Core-Sheath Fibers

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OBJECTIVE

Develop a high-throughput electrospinning process for the manufacturing of core-sheath fibers.

INTRODUCTION

Core-sheath fibers fabricated via electrospinning show great promise for use in a variety of applications including drug delivery/tissue engineering [1], self-healing coatings [2], filters [3], and super-hydrophobic materials[4]. However, needle-based, core-sheath electrospinning systems typically operate at flow rates between 1-10 ml/h, resulting in low throughput and deposition rate. Various groups have addressed this limitation by developing high throughput methods using multi-nozzles or through free liquid surface electrospinning, such as the Nanospider® developed by Elmarco [5, 6]; however, these methods are currently applicable only to monofibers. We have developed a novel electrospinning fixture capable of producing core-sheath fibers with up to 300-fold increase in volumetric throughput relative to typical needle approaches. This significant achievement in manufacturing rate will help realize the tremendous potential of core-sheath fibers.

EXPERIMENTAL

The high-throughput, needleless electrospining fixture consists of two triangular shaped troughs that are aligned to a single vertical plane to form a slit-surface (Figure 1). The core slit is set to be slightly below that of the sheath slit. Core and sheath polymer solutions are delivered to the slits through their respective fixtures by applying precise control of pneumatic pressure using syringe pumps, ensuring consistent flow rates. The fixture itself is connected to a high voltage source for generation of an electric field. We performed experiments to evaluate the effects of (1) solution flow rates and (2) solution viscosity on the formation of core-sheath Taylor cones. Table I details the polymer systems used in the experiments.

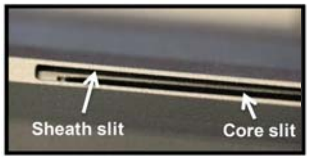

Figure 1. One-dimensional core and sheath slit-surface formed from aligning two fixtures each containing a length-wise slit.

Table I. Details of polymer systems used in the study

	Experiment 1	Experiment 2
Sheath Solution	3.5wt% PLGA in hexafluoroisopropanol	12 or 16wt% PCL in chloroform: methanol
Core Solution	12wt% PCL in 6:1 (by vol) chloroform:methanol containing 30% dexamethasone relative to PCL	

RESULTS AND DISCUSSION

Similar to a coaxial needle (which provides a single point for polymer solutions to exit), the slits of the needleless electrospinning fixture provides a line along which polymer solutions can exit and co-localize ("one-dimensional" electrospinning). Upon application of a critical electric field strength, multiple jets initiate along the length of the slit-surface as shown in Figure 2A. As the core and sheath solutions exit from their respective slits, they co-localize to form multiple core-sheath Taylor cones spontaneously that ultimately leads to core-sheath fibers (Figure 2B-D). This process occurs within a few seconds and starts with the formation of an electrospinning jet composed of the sheath polymer only (Figure 2B). We hypothesize that the internal fluid pressure will drop at the locations where sheath solution jets are present. As a result, the inner core solution will preferentially flow towards locations with lower relative pressure. Ultimately, we hypothesize that viscous shear forces of the sheath solution entrain the core solution to form a stable core-sheath Taylor cone (Figure 2D).

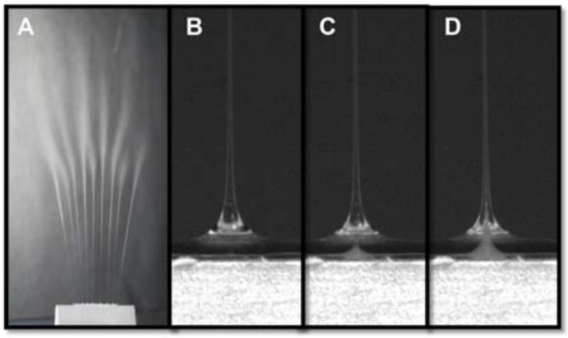

Figure 2. (A) Example of multiple electrospinning jets formed across a slit-surface. (B) Electrospinning jet formed from sheath solution without core solution entrainment. (C) Same electrospinning jet as in (B), demonstrating the spontaneous entrainment of core solution. (D) Fully formed electrospinning jet exhibiting a core-sheath structure.

Using this needle-less fixture, we have been able to operate at total flow rates up to 300 ml/h - an order of magnitude higher than our electrospinning using a needle-based system. Furthermore, preliminary data indicate that the system is scalable, thereby increasing throughput even further. To the best of our knowledge, the data presented here represent the first time that core-sheath fibers have been electrospun at such high volumes. The diameter of the core-sheath fibers produced using the polymer systems listed in Table I were around 2-4 micron, which is similar in range to what is achieved with a core/sheath needle-based systems. An example of the fibers produced along with a cross-sectional image showing the core-sheath structure is shown in Figure 3. We next performed a series of studies to identify and understand the variables and conditions under which core-sheath Taylor cones form using our novel fixture design.

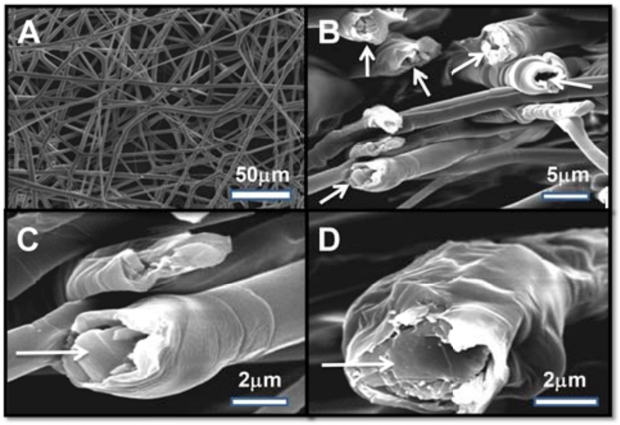

Figure 3. (A) Scanning electron micrograph of core-sheath fibers fabricated using needle-less fixture. (B) Low magnification cross-sectional image of multiple fibers showing core-sheath structure (white arrows). (C-D) High magnification image of fiber cross-section showing distinct crystalline drug core (white arrow) enclosed by sheath polymer.

Solution flow rate. The effect of solution flow rate on core-sheath Taylor cone formation was investigated by (1) keeping the sheath flow rate constant while varying the core flow rate and (2) keeping the core flow rate constant while varying the sheath flow rate. For the first experiment, the sheath flow rate was kept constant at 200 ml/h while varying the core flow rate to 20, 40, and 60 ml/h. As shown in Figure 4A-C, distinct core-sheath Taylor cones could be visualized only when the core flow rate was set to 40 or 20 ml/h (Figure 4A, B). The conditions with successful core-sheath Taylor cone formation corresponded to when the calculated total solution (core + sheath) flow velocity exiting the top of the slit was greater than the core solution flow velocity. Moreover, a greater difference between these two values resulted in a more distinct core-sheath structure.

For the second study, the sheath flow rate was varied to 200, 100, or 40 ml/h while the core flow rate was kept constant at 20 ml/h (Figure 4 D-F). The results for this set of experiments were similar to before; specifically, distinct core-sheath Taylor cones formed only when the total solution velocity was greater than the core velocity. This condition was true when the sheath flow rate was set at 100 ml/h or above (Figure 4 D, E). These results suggest that control over core-sheath Taylor cone structure can be manipulated via flow velocities of the solutions.

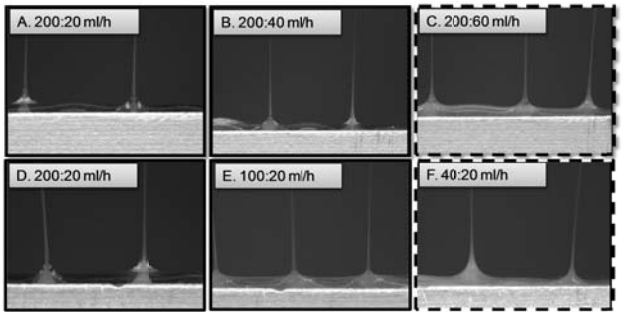

Figure 4. Example images of core/sheath Taylor cones from the flow rate study illustrating flow rate conditions in which distinct (solid border) and non-distinct (dashed border) core/sheath Taylor cones were formed. (A-C) Set of conditions in which the core flow rate was varied. (D-E) Set of conditions in which the sheath flow rate was varied.

Solution viscosity. The impact of solution viscosity on core-sheath Taylor cone formation was also investigated. As shown in Table I, either a 12wt% or 16wt% PCL solution was used as the sheath solution, resulting in differences in viscosity of 280 cP vs. 760 cP, respectively. The viscosity of the core solution was 500 cP. In this experiment, the flow rates for both systems were set at 200 and 20 ml/h for the sheath and core solutions, respectively. It was found that the core-sheath formation and morphology of the Taylor cones was more distinct when 16wt% PCL was used as the sheath solution, even though the same flow rates were used. We hypothesize that this results from a shear force sufficient to entrain the core solution due to a sheath solution viscosity higher than the core solution viscosity (760 > 500 cP). In contrast, the 12wt% PCL solution has a viscosity lower than that of the core solution (280 < 500cP) and did not exhibit distinct core-sheath Taylor cone formation. The data here indicate that flow velocity is not the only factor that determines whether core/sheath Taylor cones form, but that solution viscosities also play a role (Note: The conditions shown here meet the conditions of sheath flow velocity being greater than core flow velocity as described in the previous section).

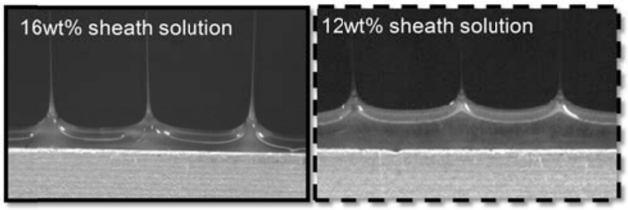

Figure 5. Images of distinct and non-distinct core/sheath Taylor cones using sheath solutions of different viscosities. Solid and dashed border indicates distinct or non-distinct core/sheath Taylor cones, respectively.

CONCLUSION

We have developed a novel needle-less electrospinning fixture capable of producing core-sheath fibers at high throughput levels. Solution velocity and viscosity are two important parameters that can be manipulated to obtain multiple core-sheath Taylor cones, jets and electrospun fibers. This technology has the potential to address the current industrial manufacturing limitations for the production of core-sheath fibers.

ACKNOWLEDGMENT

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