Core-Shell Hollow Microfibers by Triaxial Electrospinning

Abdurizzagh Khalf, Kumar Singarapu and Sundararajan V. Madihally.
School of Chemical Engineering, Oklahoma State University, Stillwater, OK.

Introduction: Electrospinning technique uses an electric field to produce nano and microfibers. Since fibers of sizes mimicking in vivo matrix configuration can be formed, electrospinning has gained significant attention in regenerative and drug delivery applications. Selecting suitable synthetic and natural polymers one can tailor the mechanical and biological properties by manipulating process parameters of electrospinning technique. Co-axial and triaxial electrospinning are modified methods for preparing fibers with multi-layered walls. Reasons for these modifications include increasing the surface area to deliver important molecules, improving the mechanical properties, and enhancing the structural morphology. In co-axial electrospinning, two different polymer solutions and in triaxial electrospinning three different polymers solutions are supplied into a compound Taylor cone (three flow streams) through a spinneret and charged liquid at the nozzle orifice is drawn out by electric field to form a liquid jet. The objective of this study was to design a triaxial electrospinning spinneret capable of producing multi wall core-shell fibers useful for biomedical applications, and to investigate the effect of solvent system properties on the fabrication of core shell hollow nanofibers made of polycaprolactone (PCL), cellulose acetate (CA), and polyvinyl alcohol (PVA).

Methods: The electrospinning setup consists of a syringe pump, syringes, spinneret, high voltage power supply, earth grounding, and a collector plate. Spinneret design (Figure 1a) consisted of a plastic tee fitted with two concentric needles modified to create three streams flow. PCL (Mw = 85kDa), PVA (99% hydrolyzed, 110 kDa) and CA (30kDa) were mixed in chloroform and methanol (9:1 v/v) solvent system at 20% mass concentration, acetone and dioxin (2:1 v/v) at 14 wt %, and distilled water under heating at 80°C respectively. Electrospinning was performed using mineral oil (99.9%) as the inner core and removed by submersing the fibers in octane for over a period of 24h. Formed fibers were analyzed using a scanning electron microscope. To locate the distribution of the polymer, one of them was stained with Rhodamine B prior to electrospinning and formed fibers were analyzed using fluorescence microscopy. Formed scaffolds were also seeded with human umbilical vein endothelial cells (HUVEC) and evaluated for spreading and toxicity. All HUVECs were pre-stained with CFDA-SE prior to seeding. Also, scaffolds with cells were assessed by SEM.

Results: When PCL was the outer shell and CA was the inner shell, uniform microsize fibers were formed without difficulty (**Figure 1b**). Similar to single fiber formation, nanopores were observed in the outer shell when fibers were formed under high humidity conditions. Next, we questioned the ability to switch the configuration to CA outershell and PCL inner shell. Switching the orientation of solutions did not result in uniform fiber formation. To understand the rationale, we assessed the boiling point of the solvents using a ChemCAD simulator and obtained the vapor-liquid equilibrium diagrams. These results showed that the boiling point of PCL solvent was lower than that of CA solvent. Thus, rapid solvent evaporation of the inner shell caused phase separation, leading to failure in obtaining fibers. We increased the boiling point of PCL solvent by adding acetic acid. Upon changing the PCL solvent, uniform fibers were formed and analysis of CA distribution showed its presence in the outer shell (**Figure 1c**). Since PVA is water soluble, we tested the ability to form PCL outer shell and PVA inner shell configuration. Uniform fibers were formed. However, when the configuration was changed, fibers did not form. To change the boiling point of PVA solvent, we tried to add methanol, acetone, and other solvents. However, presence of other solvents prematurely precipitated PVA. Electrospinning also showed a similar effect. Thus, apart from boiling point of solvent, polymer solubility is also important. Next we assessed the HUVEC behavior on these scaffolds (**Figure 1d**). HUVECs showed cell spreading similar to that on the tissue culture plastic and the viability was similar to that of TCP. This suggested that the formed scaffolds are not toxic.

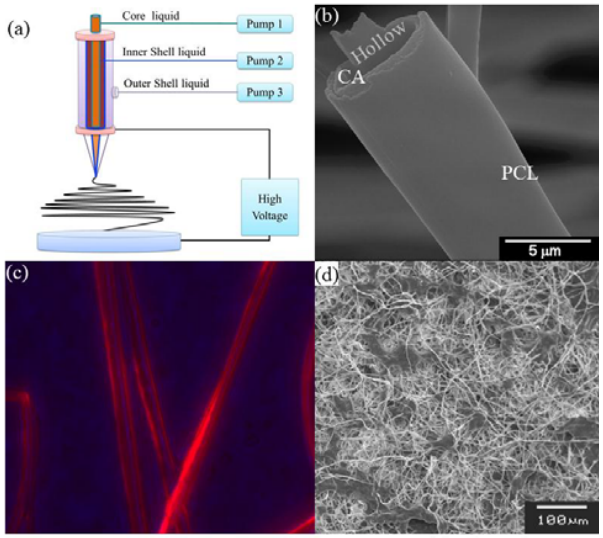

Figure. Triaxial spinning of PCL, PVA and CA. (a) schematic showing the spinneret design. (b) Micrograph showing outer PCL, and inner CA fibers. (c) Fluorescent micrograph of Rhodamine B stained CA, PCL hollow fiber. (d) Micrograph showing HUVEC distribution on CA-PCL hollow fibers after 24 h.

Conclusions: Hollow fibers of PCL-CA, CA-PCL and PCL-PVA were produced by triaxial electrospinning. It is important to have a solvent system in the outer shell that has a boiling point less than that in the inner shell. Also, solubility of the polymers in the presence of solvents used in dissolving other polymers is important. Formed fibers were not toxic to endothelial cells. Using this technique, fibers can be formed with unique properties for use in various biomedical applications.

Acknowledgments: Financial support from OCAST (HR12-023) and Edward Joullian Endowment.