NUMERICAL SIMULATION STUDY OF A STABLE JET SHAPE VARIATION IN ELECTROSPINNING

by

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Original scientific paper
https://doi.org/10.2298/TSCI170615191W

High voltage power was used to produce jet in electrospinning. It was very difficult to study electrospinning jets by experiment, because they have high-speed and complex movements in the high-voltage electrostatic field, and the diameter of jet was very small. In this study, the software of finite element analysis was used to simulate the formation process of a stable jet in electrospinning. The numerical simulation results indicated that the diameter of a stable jet decreased as well as the velocity of a stable jet increased with the increasing of drafting force when the solution flow rate was constant. At last, an experiment about a stable jet diameter has been carried out. The different conductivities spinning solution by adding different content lithium chloride into polyvinyl alcohol solution have been prepared. They could lead to different electric force for a stable jet in electrospinning. We used glass slide to intercept the stable jet to test the diameter of jet. The experiment results showed that the diameter of a stable jet decreased with the electric force increased. The experiment results were in good agreement with numerical simulation of a stable jet in electrospinning.

Key words: *numerical simulation, electrospinning, jet, electric force, diameter*

Introduction

Electrospinning technology is an effective and convenient method to fabricate ultrafine fiber from submicron fiber to nanofiber. Electric force plays an important role in the formation and movement of jet during electrospinning. When electric force overcomes the solution surface tension, Taylor cone is formed in the tip of nozzle [1, 2]. The jet is elongated under the effect of electric force, the jet has first a short stable section and then whipping section quickly, the solvent evaporates and then solidified nanofibers are collected on a plate [3]. A conventional electrospinning set-up consists of three parts: the solution supply system, the nanofiber collection system, and the high voltage power supply. The schematic diagram of electrospinning is showed in fig. 1.

It is very significant for us to understand and investigate the formation and movement process of electrospinning jet, this instructs us to choose spinning process parameter and improve quality of product. There are many references about the formation mechanism of electrospinning [4-10]. Spivak and Dzenis [11] derived a differential equation for the variation of jet radius with axial co-ordinate and analyzed asymptotic variation of the jet radius at large distances from the jet origin. Carroll and Joo [12] investigated the modeling and simula-

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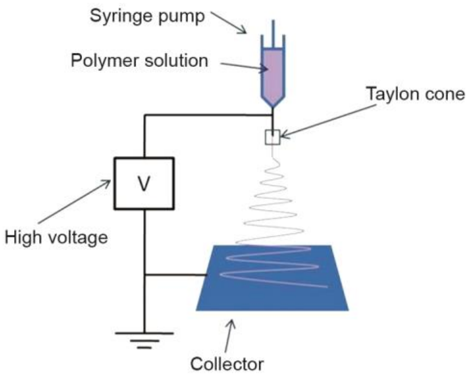

Figure 1. The conventional electrospinning set-up

tion of the initial stable jet phase seen during the electrospinning process. The conductivity of solution could be changed by adding lithium chloride. The theoretical analysis showed that the relationship between radius, r, of jet and the axial distance, z, from nozzle follows an allometric law in the form $r \sim z^{-0.5}$ in the case of surface electric charge [13]. With the development of numerical compute technology, more and more researchers adopt finite element analysis software to investigate the formation process of jet because it can mimic experiment process and improve research efficiency. Much works have been conducted about numerical simulation in electrospinning. Xu *et al.* [14] investigated numerical simulation of the two-phase flow in the single-bubble electrospinning process by using the volume of fluids method. The results showed that the numerical simulation results were well consistent with the experimental data. The motion of the jet and the distribution of jet velocity also have been simulated and discussed using FLUENT software [15]. In addition, a discrete mathematical model about the magnetic electrospinning jet was established to investigate the effect of excitation current on electrospinning instability, the simulation results agree well with the experiment data [16, 17]. A discrete model was used to simulate the bending instability phenomenon in electrospinning, the simulation results showed that the shape of the instability region was an expanding spiral [18]. Hu and Huang [19] proposed a mathematic physical model to study two-phase flows occurring in coaxial electrospinning. Wei *et al.* [20] studied a physical model of electric field induced by charged droplets, the diameter of drops were predicted at various applied voltages and flow rates, the results showed that the simulation calculated droplet diameter agrees well experimental measurement. Electrospinning jets often involve two-phase flow, the formation and movement of jets is very complex mechanism. It is convenient to observe and analyze the variation process of jet with the aid of CFD. Numerical simulations can be used to investigate aspects of the mechanism that are not amenable to experiment. Some researchers used CFD and experimental analysis to present a new approach about optimizing a double-channel pump [21]. The CFD was adopted to analysis and design optimization of jet pumps [22]. The CFD also was used to conduct numerical assessment of ejector operation for refrigeration applications [23].

In this paper, we used ANSYS 14.5 to simulate the shape variation of jet under the effect of different electric force based on a slender-body physics model. It is very difficult to test and discuss the jet formation and the electric force during electrospinning experiment. The jet shape variation process could be observed clearly with different drafting force in the contours of numerical simulation. Experiment has been performed to measure the diameter of jet using the glass slide intercept method. The results showed that the numerical simulation results of jet shape variation for stable section agreed quite well with the experimental results. The numerical simulation method could provide an effective way to understand the shape variation process for a stable jet during electrospinning.

Numerical simulation

Theoretical analysis for a stable jet

It was very important to understand stress conditions of a stable jet before numerical simulation. In here, the classical slender-body model for jet was introduced to analysis a stable jet [24]. The schematic diagram for a short segment force analysis of a stable jet was shown in fig. 2(a). In the part of the stable jet, the jet was stretched by the electric force and gravity. The momentum equation of was established by considering the forces on a short segment of the stable jet of electrospinning in 3-D.

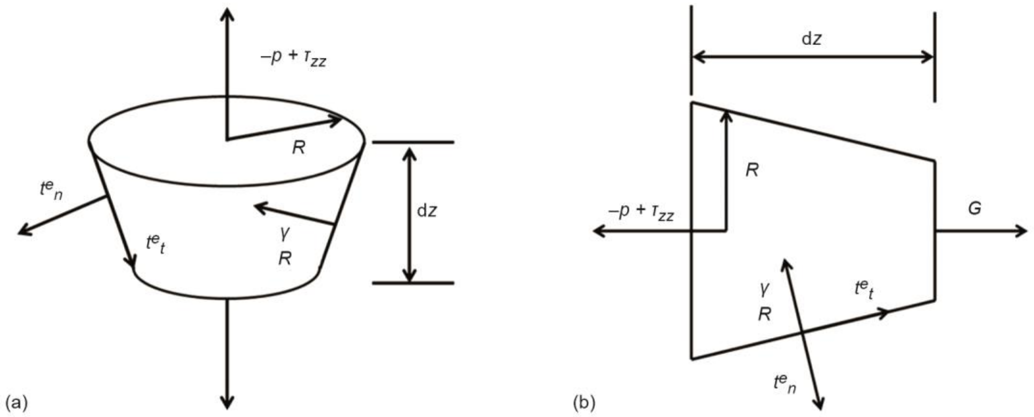

Figure 2. Force analysis on short segment for a stable jet in (a) 3-D, (b) 2-D

The momentum equation of Slender-Body Model could be expressed [24]:

$$\frac{\mathrm{d}}{\mathrm{dz}}(\pi R^2 \rho \upsilon^2) = \pi R^2 \rho \mathrm{g} + \frac{\mathrm{d}}{\mathrm{dz}}[\pi R^2(-p + \tau_{zz})] + \frac{\gamma}{R} 2\pi R R^* + 2\pi R(t_\mathrm{t}^e - t_\mathrm{n}^e R^*) \tag{1}$$

where τ_{zz} is the axial viscous normal stress, p – the inner pressure, ρ – the density of solution, υ – the velocity of stable jet, γ – is the surface tension, g – the gravity acceleration, t_t^e and t_n^e are the tangential and normal tractions on the surface of the jet, respectively, z – the length of stable jet along axis direction, and R^* – the slope of the jet surface.

However, because of the complexity of force analysis and momentum equation for a stable jet in 3-D, it was restrained to use it to solve practice problems. Therefore, we developed 2-D force analysis model for a stable jet to extend its practice applications based on 3-D. For a stable jet in 2-D, some forces could be simplified. Force analysis on a short segment of a stable jet in 2-D was shown in fig. 2(b). Since the jet was mainly stretched by the electric force and gravity, the force caused by the surface tension and normal electric tractions could be missed. Balance equation could be expressed:

$$\frac{\gamma}{R} 2\pi R R^* - 2\pi R t_\mathrm{n}^e R^* \approx 0 \tag{2}$$

The momentum equation of could be modified:

$$\frac{\mathrm{d}}{\mathrm{dz}}(\pi R^2 \rho \upsilon^2) = \pi R^2 \rho \mathrm{g} + \frac{\mathrm{d}}{\mathrm{dz}}[\pi R^2(-p + \tau_{zz})] + 2\pi R t_\mathrm{t}^e \tag{3}$$

While the tangential stress in the axial direction it becomes $t_{\mathrm{t}}^{e}\cos\theta$, where θ is the separation angle between the axial direction and the direction of t_{t}^{e}. Because of the changes in diameter along the jet axis which we could almost ignore, θ is close to 0°. Thus we could still assume that $t_{\mathrm{t}}^{e}\cos\theta \approx t_{\mathrm{t}}^{e}$. Due to the diameter of a stable jet was very thin, the effect of gravity could be ignored. Finally, the drafting force of a stable jet in the axial direction could be expressed:

$$f_n = \pi R^2(-p+\tau_{zz}) + 2\pi R t_{\mathrm{t}}^{e} \tag{4}$$

The drafting force, f_n, is the resultant force caused by the axial viscous normal, inner pressure and the tangential electric tractions force.

Mathematical model

In the process of electrospinning, a stable jet was pulled from a needle tip and accelerated by a constant external electric field. Considering the gravity force, viscous force and electric force, constitutive relations of mass conservation equation, momentum conservation equation, and electric charge conservation equation were expressed [20, 25]:

$$\nabla\upsilon = 0 \tag{5}$$

$$\frac{\partial(\rho\upsilon)}{\partial t} + \nabla(\rho\upsilon\upsilon) = -\nabla p + \nabla(\sigma^{\mathrm{f}} + \sigma^{\mathrm{e}}) + f_{\mathrm{b}} \tag{6}$$

$$\nabla J = 0 \tag{7}$$

where ρ is the solution density, υ – the solution velocity, t – the time, p – the pressure, σ^{f} – the viscous force, σ^{e} – is the electric force, and f_{b} – the body force.

The viscous force, σ^{f}, the electric force, σ^{e}, and the electric body force, f_{e} were expressed [20]:

$$\sigma^{\mathrm{f}} = \mu[\nabla\upsilon + (\nabla\upsilon)^T] - \frac{2}{3}\mu(\nabla\upsilon) \tag{8}$$

$$\sigma^{\mathrm{e}} = \varepsilon\varepsilon_0 EE - \frac{\varepsilon\varepsilon_0}{2}EE\left(1 - \frac{\rho}{\varepsilon\varepsilon_0}\frac{\partial\varepsilon\varepsilon_0}{\partial\rho}\right) \tag{9}$$

$$f_{\mathrm{e}} = \nabla\sigma^{\mathrm{e}} = qvE - \frac{1}{2}E^2\nabla\varepsilon\varepsilon_0 + \nabla\left(\frac{1}{2}\rho\frac{\partial\varepsilon\varepsilon_0}{\partial\rho}E^2\right) \tag{10}$$

where J is the identity tensor, μ – the dynamic viscosity, ε – the electric permittivity, $\varepsilon_0 = 8.85\cdot10^{-12}$ [$\mathrm{CV^{-1}m^{-1}}$] – the permittivity vacuum, qv – the volumetric electric charge density, and E – the electric field intensity.

Physical model of a stable jet

The ANSYS 14.5 workbench contained four part models which were fluids, structures, electronics and systems. In this study, we used POLYFLOW software which belonged to fluids model. Physical model of a stable jet was built based on practice a stable jet size. The length of geometry was $8\cdot10^{-3}$ m and the width of geometry was $5\cdot10^{-4}$ m. The shape of geometry was rectangle. Geometric model of a stable jet could be seen in fig. 3(a). The line of ac was inlet and the line of bd was outlet. The two lines of ab and cd represented walls. The stable jet has been considered as viscoelasticity model in POLYFLOW software, computational domain sizes were geometry of a stable jet. Boundary conditions: The inlet velocity was set as solution flow rate, the outlet was applied different drafting force f_n, two walls were set free surfaces. Mesh of geometric could be automatically generated by POLYFLOW software. The quality of mesh has an influence on the precision and efficiency in simulation computation. Mesh model of a stable jet was shown in fig. 3(b). In the process of numerical simulation, the relevant parameters were listed: the material was polyvinyl alcohol solution which belonged to viscoelasticity fluid, the viscosity of solution was 47 Pa·s, the density was 1000 kg/m^3, the inlet velocity was $2.22 \cdot 10^{-4}$ mL/s.

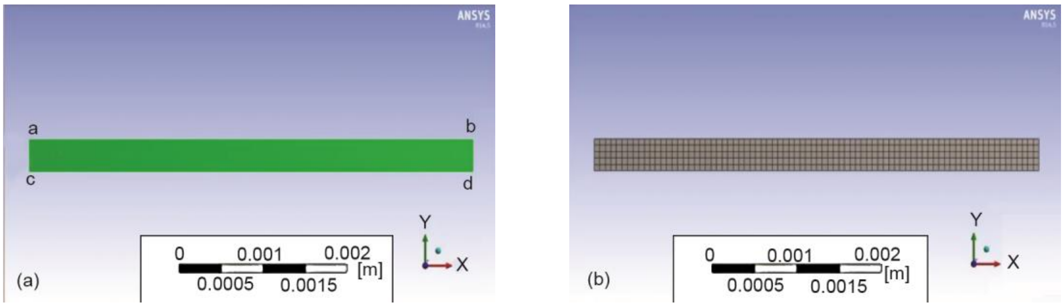

Figure 3. (a) Geometric model of a stable jet, (b) mesh model of a stable jet
(*for color image see journal web site*)

Generalized Newtonian isothermal flow problem was used to solve physical model of a stable jet in POLYFLOW software.

Numerical simulation results and analysis

The shape variations of the stable jet with different axial drafting force were shown in fig. 4. The blank arrow direction represented the direction of drafting force. The drafting force was the resultant force caused by viscosity force and electric force. As to the certain polymer solution, the viscosity force was constant. The drafting force depended on the electric force. The changes of the drafting force meant the variations of the electric force. It could be seen that six drafting forces has been chose to represent different electric forces. In fact, those six drafting forces were not the true force value in practice situation. In order to simulate the variation of a stable jet with different electric force, those six drafting force could be set through simulation software. From fig. 4, we could see that the diameter of a stable jet decreased gradually along axis distance of a stable jet. In addition, the diameter of a stable jet decreased gradually with the increasing of different drafting forces. Some reports found the electric field and the solution speed between the nozzle and the collection device affect the shape of the spinning jet in electrospinning [26]. Some other researchers argued that with the electric field enhancing, the jet ejected from the Taylor cone would thin more rapidly, and the profile of the jet at the top of the Taylor cone would become shorter [9]. In this study, the results of a stable jet simulation with different drafting force validated their experimental results.

The velocity curves of stable jet with different drafting forces along axis distance were shown in fig. 5. When the drafting force was small, the velocity of stable jet was basically a contant value of about 0.444 m/s. The velocity of stable jet increased gradually with the increasing of drafting force, the slope of the velocity curves also increased gradually. When drafting force reached to a maximum value, the velocity value of stable jet could reach 0.7978 m/s. The numerical simulation results showed that the velocity of a stable jet has increased with the increasing of the electric force. The diameter of a stable jet has decreased with the increasing of electric force. The simulation velocity results of the stable jet were consistent with the experiment results of previous study [1]. We could make full use of the software of numerical simulation to study the movement mechanism of jet in electrospinning.

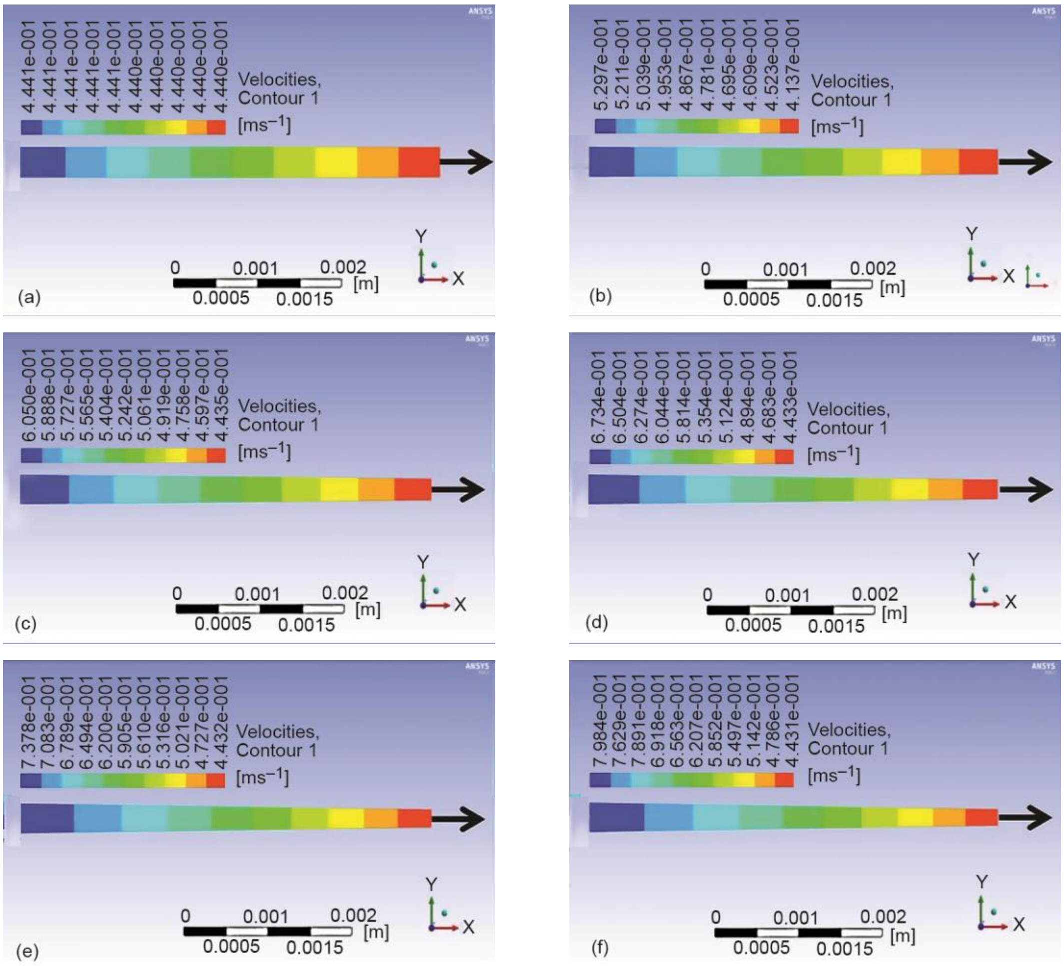

Figure 4. The shape variations of the stable jet with different axial drafting force of (a) f_n = 5, (b) f_n = 2214, (c) f_n = 4423, (d) f_n = 6632, (e) f_n = 8841, (f) f_n = 11050 (*for color image see journal web site*)

Experimental details

Materials

Polyvinyl alcohol (PVA, Mn = 88,000 g/mol) was obtained from J & Kchemica®. Distilled water was used as the solvent. Lithium chloride (LiCl) was purchased from the Pinjiang Chemical Co. Ltd. Different concentration LiCl (0.1 wt.%, 0.2 wt.%, 0.3 wt.%, 0.4 wt.%, 0.5 wt.%, and 0.6 wt.%) were added to distilled water and the solution was stirred until the LiCl was dissolved completely. Then PVA polymer was added to the solution incrementally until it reached 14 wt.% solution of PVA polymer in distilled water. The solution was stirred for 4 hours at 80 °C to obtain an homogenous polymer solution.

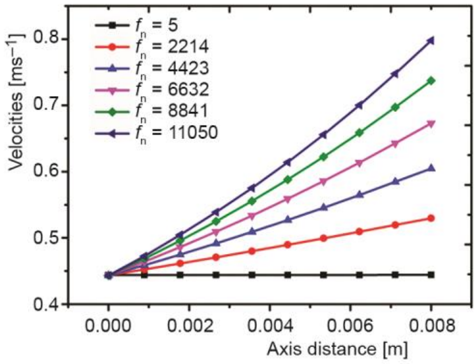

Figure 5. The velocity curves of stable jet with different drafting forces along axis distance

Electrospinning set-up

A conventional single needle electrospinning set-up was used to carry out experiment. The applied voltage was 20 kV. The distance between the needle and the collector was 15 cm and the diameter of the needle was 1 mm. The solution flow rate was 0.8 mL per hour. A schematic diagram of intercepting jet using glass slide was shown in fig. 6. The experiment was performed under ambient conditions at room temperature 20 °C and relative humidity of about 50%. The conductivities of adding different concentration LiCl spinning solutions were tested by conductivity meter. The shape variation of a stable jet could be observed in optical microscope.

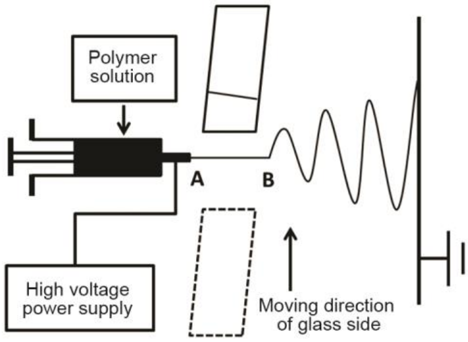

Figure 6. The schematic diagram of intercepting jet using glass slide

Results and discussion

Table 1 shows comparison of conductivity of solutions with different concentrations of LiCl. We could see that the conductivities increased with the increasing of adding LiCl concentrations. When the concentration of LiCl was 0.1 wt.%, the conductivity of solution only was 2376.33 μs/cm. However, the concentration of LiCl was 0.6%, the conductivity of solution could reach 4957.99 μs/cm. The results showed a linear increase relationship between the concentration of LiCl and its conductivity with certain range. The charge density of jet increased with the increasing of solution conductivity. This could lead to the increasing of electric force. The relationship between the axis distance and the diameter of jet was obtained by using glass slide to intercept jet. The diameter of jet at different points corresponding different axis distances were measured by the optical microscopy. The shape variation of stable jet observed in the optical microscope was shown in fig. 7.

Table 1. Comparison of conductivity of solutions with different concentrations of LiCl

Concentration of LiCl [wt.%]	0.1	0.2	0.3	0.4	0.5	0.6
Conductivity [μscm^{-1}]	2376.33	2892.67	3408.99	3925.33	4441.67	4957.99

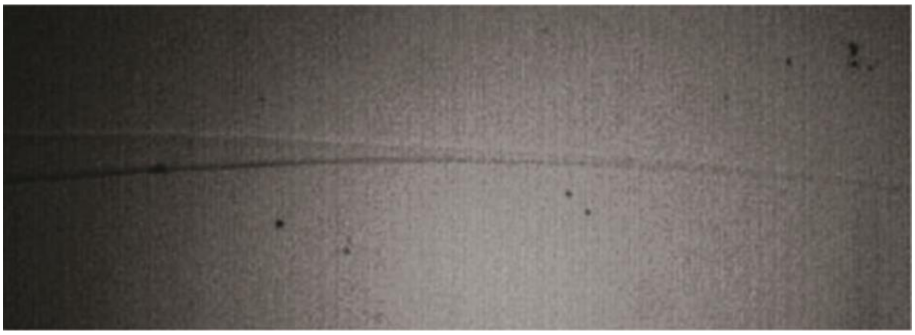

Figure 7. The shape variation of a stable jet observed in the optical microscope

As was known to all, electric force played an important role in the process of jet formation for electrospinning. Electric force exceeded the resultant force from viscosity force and surface tension which could lead to form jet. After the jet formation, the drafting force of stable jet received was the resultant force caused by viscosity force and electric force. The drafting force increased with the increasing of electric force based on eq. (4). Adding salt into the polymer solution could improve the conductivity of solution. The surface charge density of stable jet increased and the Coulomb force of the stable jet increased. The electric force of the stable jet received increased with the increasing of solution conductivity. In order to verify the numerical simulation results, the different concentrations of LiCl were added into the polymer solution of PVA to change the different electric forces of jet received based on the relationship between the electric force and the conductivity. The relationship between the stable jet diameter and axis distance with different LiCl concentrations has been seen in fig. 8. The results showed that the diameter of the stable jet decreased gradually with the increasing of axis distance. The diameter of the stable jet also decreased with the increasing of different LiCl concentrations from 0.1 wt.% to 0.6 wt.%. The reason was attributed to the increasing of the conductivity, that could led to the increasing of electric force, so the drafting force of the stable jet received increased which resulted from the decreasing of the stable jet diameter. The same results could be seen in numerical simulation, the stable jet diameter decreased with the increasing of simulation drafting force, at the same time, the velocity of stable jet increased. The results showed that experiment results were in good agreement with numerical simulation results.

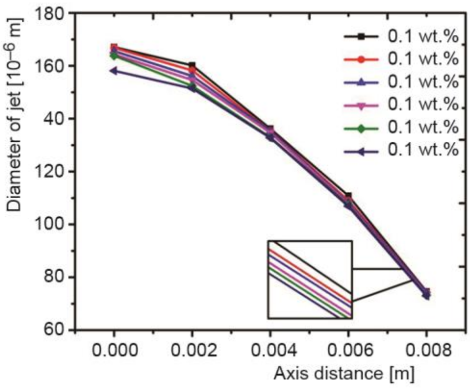

Figure 8. The relationship between the stable jet diameter and axis distance with different LiCl concentrations

Conclusion

The 2-D physical model of the stable jet was established based on the slender-body model in the electrospinning. The mathematical model of the stable jet was established using mass conservation equation, momentum conservation equation, and electric charge conservation equation. The numerical simulation of stable jet was carried out by using the software of finite element analysis. The diameter variations contours and velocity curves of stable jet were obtained through numerical simulation. The numerical simulation results showed that the stable jet diameter became thinner and the velocity of stable jet increased with the increasing of drafting force. In order to verify the results of the numerical simulation, the different concentrations for LiCl were added into the polymer solution of PVA to change the conductivity of solution to represent the different electric force based on the relationship between conductivity and electric force. The drafting force increased due to the increasing of electric force. The experimental results indicated that the stable jet diameter decreased gradually with the increasing of concentrations LiCl. The experimental results were in good agreement with the numerical simulation results. Therefore, it was very convenient and clear to analyze and study the shape variation and movement of stable jet in electrospinning using the numerical simulation method.

Acknowledgment

This work was partly supported by the Chang Jiang Youth Scholars Program of China and grants (51373033 and 11172064) from the National Natural Science Foundation of China to Prof. Xiaohong Qin. As well as *The Fundamental Research Funds for the Central Universities* and *DHU Distinguished Young Professor Program* to her. It also has the support of the Key grant Project of Chinese Ministry of Education.(No 113027A). This work has also been supported by *Sailing Project* from Science and Technology Commission of Shanghai Municipality (14YF1405100) to Dr. Hongnan Zhang. This work was also supported by the Fundamental Research Funds for the Central Universities (CUSF-DH-D-2016008) to Mr. Liang Wei.

References

[1] Reneker, D. H., Yarin, A. L., Electrospinning Jets and Polymer Nanofibers, *Polymer*, *49* (2008), 10, pp. 2387-2425
[2] Yarin, A. L., *et al.*, Bending Instability in Electrospinning of Nanofibers, *Journal Applied Physics*, *89* (2001), 5, pp. 3018-3026
[3] Deitzel, J. M., *et al.* Controlled Deposition of Electrospun Poly(Ethylene Oxide) Fibers, *Polymer*, *42* (2001), 19, pp. 8163-8170
[4] Yarin, A. L., *et al.*, Taylor Cone and Jetting from Liquid Droplets in Electrospinning of Nanofibers, *Journal of Applied Physics*, *90* (2001), 9, pp. 4836-4846
[5] Reneker, D. H., *et al.*, Bending Instability of Electrically Charged Liquid Jets of Polymer Solutions in Electrospinning, *Journal of Applied Physics*, *87* (2000), 9, pp. 4531-4547
[6] Yarin, A. L., *et al.*, Bending Instability in Electrospinning of Nanofibers, *Journal of Applied Physics*, *89* (2001), 5, pp. 3018-3026
[7] Reneker, D. H., Yarin, A. L., Electrospinning Jets and Polymer Nanofibers, *Polymer*, *49* (2008), 10, pp. 2387-2425
[8] Hohman, M. M., *et al.*, Electrospinning and Electrically Forced Jets. I. Stability Theory, *Physics of Fluids*, *13* (2001), 8, pp. 2201-2220
[9] Hohman, M. M., *et al.*, Electrospinning and Electrically Forced Jets. II. Applications, *Physics of Fluids*, *13* (2001), 8, pp. 2221-2236
[10] Shin, Y. M., *et al.*, Experimental Characterization of Electrospinning: The Electrically Forced Jet and Instabilities, *Polymer*, *42* (2001), 25, pp. 9955-9967
[11] Spivak, A. F., Dzenis Y. A., Asymptotic Decay of Radius of a Weakly Conductive Viscous Jet in an External Electric Field, *Applied Physics Letters*, *73* (1998), 21, pp. 3067-3069
[12] Carroll, C. P., Joo, Y. L., Discretized Modeling of Electrically Driven Viscoelastic Jets in the Initial Stage of Electrospinning, *Journal of Applied Physics, 109* (2011), 9, 094315

[13] Qin, X. H., *et al.*, Effect of LiCl on Electrospinning of PAN Polymer Solution: Theoretical Analysis and Experimental Verification, *Polymer*, *45* (2004), 18, pp. 6409-6413
[14] Xu, L., *et al.*, Numerical Simulation for the Single-Bubble Electrospinning Process, *Thermal Science*, *19* (2015), 4, pp. 1255-1259
[15] Xu, L., *et al.*, Numerical Simulation of a Two-Phase Flow in the Electrospinning Process, *International Journal of Numerical Methods for Heat & Fluid Flow*, *24* (2014), 8, pp. 1755-1761
[16] Xu, L., *et al.*, Numerical Study of Magnetic Electrospinning Processes, *Computers & Mathematics with Applications*, *61* (2011), 8, pp. 2116-2119
[17] Xu, L., A Mathematical Model for Electrospinning Process under Coupled Field Forces, *Chaos Solitons & Fractals*, *42* (2009), 3, pp. 1463-1465
[18] Zeng, Y. C., *et al.*, Numerical Approach to Electrospinning, *International Journal of Nonlinear Sciences and Numerical Simulation*, *7* (2006), 4, pp. 385-388
[19] Hu, Y., Huang, Z. M., Numerical Study on Two-Phase Flow Patterns in Coaxial Electrospinning, *Journal of Applied Physics*, *101* (2007), 8, 084307
[20] Wei, W., *et al.*, Numerical Simulation of the Cone-Jet Formation and Current Generation in Electrostatic Spray-Modeling as Regards Space Charged Droplet Effect, *Journal of Micromechanics and Microengineering*, *23* (2013), 1, pp. 57-78
[21] Zhao, B. J., *et al.*, Optimization Design of a Double-Channel Pump by Means of Orthogonal Test, CFD, and Experimental Analysis, *Advances in Mechanical Engineering*, *6* (2014), July, pp. 1-10
[22] Fan, J., *et al.*, Computational Fluid Dynamic Analysis and Design Optimization of Jet Pumps, *Computers & Fluids*, *46* (2011), 1, pp. 212-217
[23] Aidoun, Z., *et al.*, Numerical Assessment of Ejector Operation for Refrigeration Applications Based on CFD, *Applied Thermal Engineering*, *26* (2006), 5-6, pp. 604-612
[24] Feng, J. J., The Stretching of an Electrified Non-Newtonian Jet: A Model for Electrospinning, *Physics of Fluids*, *14* (2002), 11, pp. 3912-3926
[25] Spivak, A. F., *et al.*, A Model of Steady State Jet in the Electrospinning Process, *Mechanics Research Communications*, *27* (2000), 1, pp. 37-42
[26] Shin, Y. M., *et al.*, Experimental Characterization of Electrospinning: The Electrically Forced Jet and Instabilities, *Polymer*, *42* (2001), 25, pp. 9955-9967

Paper submitted: June 15, 2017
Paper revised: July 20, 2017
Paper accepted: August 13, 2017