Two-Dimensional Simulation of Hollow Fiber Membrane Fabricated by Phase Inversion Method

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ABSTRACT: In the steady fabricating process, two-dimensional hollow fiber membrane near the spinneret was numerically simulated using the finite element method (FEM). The unknown positions of free surface and moving interface were calculated simultaneously by the velocity and pressure fields. The effects of seven relevant parameters, i.e., inertia term, gravity term, dope flow rate, bore flow rate, dope viscosity, tensile force, end velocity and non-Newtonian on the velocity and diameter profile were studied. On the basis of the simulated results, the inertia term in hollow fiber-spinning process was safely neglected in low speed, while the effect of gravity was not be neglected. Besides, the outer diameter of the fibers increased with an increase of dope flow rate and bore flow rate; Large tensile force or large end velocity could cause large deformation in the air gap; larger viscous dope solution tended to make less deformation in the air gap. It was found that an increase of the dope flow rate at small dope flow rate resulted in an increase of the inner diameter, while at large dope flow rate, it decreased. © 2006 Wiley Periodicals, Inc. J Appl Polym Sci 100: 2067–2074, 2006

Key words: two-dimensional simulation; hollow fiber membrane; membrane formation; phase inversion method

INTRODUCTION

Generally compared with flat membranes, hollow fiber membranes have many unique properties and have found numerous applications as well. For example, hollow fiber can provide great contact surface in small volume. Usually, polymeric hollow fibers are fabricated by two means: one is melt-spinning and the other is solution-spinning.[1] Compared with melt-spinning, polymer, solvent, and nonsolvent are included in solution spinning. There are more parameters that people can control to get the desired morphology of the hollow fiber. Naturally, complication is concomitant with more free parameters.

Steady extrusion through annular die had application in manufacturing of hollow fibers, pipes and is closely related to the film blowing and wire coating processes. Hence, the simulation of the annular extrusion process has been the subject of quite a few publications.[2] To our knowledge, in all these one-dimensional or two-dimensional studies, they did not face the challenge of moving interface, because in their studies, bore fluid is not included, or their simulation is one-dimensional. Kase and Matsuo[3,4] generalized a numerical analysis of the melt-spinning process: their work was done by solving the thin-filament equation, which were formulated by averaging the sets of fundamental governing equations over the cross-section of running filament with the appropriate empirical relations for physical properties and the transport coefficients. Freeman et al.[5] obtained the velocity and temperature profile of the drawing hollow tube using finite-element and asymptotic analyses. In their study, hollow shape was gained by imposing an internal pressure in the tube. They also concluded that thin-filament equations are not valid in the region of extrudate swell near the spinneret, but finite-element can be used to study the flow in that region, and the two methods give comparable results in the region of overlap. Lipscomb[6] discusses the sensitivity of the fiber-spinning equations to material property and process variations, using a novel numerical procedure under thin-filament limit. Chung and Xu[7]derived the basic equation to relate the velocity profile of a nascent hollow fiber in the air-gap region. Also, two simplified equations were also derived to predict the inner and outer diameters of hollow fibers. Fadhel and Xu[8] developed a numerical simulation model based on the previous one and investigated the effect of mass transfer, surface tension, and drag forces on the velocity distribution. Dpto et al.[9] derived the leading-order fluid dynamics equations of isothermal, axisymmetric,

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Newtonian, hollow, compound fibers at low Reynolds numbers. Two-dimension is necessary in simulation if die swelling and moving interface is considered. In 1998, Kostas Housiadas et al.[10] solved the moving boundary problem by mapping the inner and the outer interface of the film in the radial direction onto fixed ones. And, the transformed governing equations are solved by perturbation expansion. Tae et al.[11] studied profile development and dimensional change in the melt-spinning process of hollow fibers using a finite element method and compared a numerical simulation with experimental results. In 2000, Kostas et al.[2] studied the steady extrusion of a Newtonian liquid through an annular die and its development outside and away from the die under the influence of gravitational and surface tension force. In their studies, the finite element method is used for the simulations, and Newton–Raphson iterative scheme is employed for calculating the unknown inner and outer free surface.

The objective of this study is to solve numerically the two-dimensional, steady and Newtonian or non-Newtonian (without elastic) annular spinning flow and study the effects of dope flow rate, bore fluid rate, viscosity, take up velocity, tensile force, gravity and inertia term on the diameter and velocity profiles of the fibers. Die swell phenomena are also investigated.

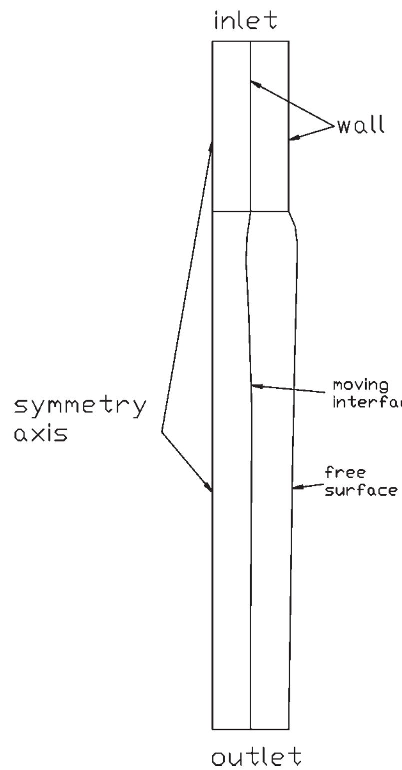

Figure 1 Geometry and boundary condition of hollow fiber in two-dimensional model.

MODEL DESCRIPTION

During the dry/wet hollow fiber spinning, strong nonsolvent (usually water) is chosen as bore fluid. The reason for the choice is that strong nonsolvent has the ability to coagulate the polymer solution at the exit of the spinneret; therefore, a thin membrane layer will be formed between the outer polymer solution and inner bore fluid, otherwise the fiber is easily broken and the polymer solution will go down as liquid drop under the force of gravitation. Assume as follows:

1. Solvent and nonsolvent mass transfer between polymer solution and bore fluid is strongly decreased by the layer. Thus, the properties of these two fluids stay as constant during the air gap distance.
2. There is no mass transfer at outer surface. This assumption is valid if high boiling solvent was used for the dope solution and if the spinning was conducted at a low temperature with relatively short air-gap distance.
3. The flow is also assumed to be isothermal and axisymmetric and is incompressible. That is

$$v_r = v_r(r,z)$$

$$v_z = v_z(r,z)$$

4. The bore fluid is Newtonian and has constant viscosity ($\mu = 1cp$). When the polymer solution is not considered as Newtonian, power law is used to describe its rheological behavior.[12]

$$\eta = m\dot{\gamma}^{n-1} \tag{3}$$

Here, m,n are constants characterizing the fluid, and $\dot{\gamma}$ is the magnitude of the rate-of-strain tensor.

5. The flow geometry and the boundary of the spinning flow are depicted in Figure 1. Because of axial symmetry, only half is presented. For convenience, cylindrical coordinates (r,θ,z) is used during the simulation. Then, the differential equations of continuity and motion for the polymer solution can be described as follows.[7,13,14]

Continuity equation:

$$\frac{1}{r}\frac{\partial}{\partial r}(rv_r) + \frac{\partial v_z}{\partial z} = 0 \tag{4}$$

Conservation of momentum:

$$r \text{ component: } \rho_h\left(v_r\frac{\partial v_r}{\partial r} + v_z\frac{\partial v_r}{\partial z}\right) = -\frac{\partial p}{\partial r} + \frac{1}{r}\frac{\partial}{\partial r}(r\tau_{rr}) + \frac{\partial \tau_{rz}}{\partial z} \tag{5}$$

$$z \text{ component: } \rho_h\left(v_r\frac{\partial v_z}{\partial r} + v_z\frac{\partial v_z}{\partial z}\right) = -\frac{\partial p}{\partial z} + \frac{1}{r}\frac{\partial}{\partial r}(r\tau_{rz}) + \frac{\partial \tau_{zz}}{\partial z} + \rho_h g \quad (6)$$

In eqs. (4)–(6), ρ_h is density of polymer solution, v_r,v_z are the velocity of r and z-components of hollow fiber, respectively. g denotes the gravitational constant, p is the isotropic pressure, and τ_{ij} is the extra stress. If tensile force is imposed, f term is added to the right side of z component of momentum conservation equation. These three equations are also true for the bore fluid.

The boundary conditions are as follows:

1. Inlet condition: The flow is fully developed that all derivatives normal to the inlet are zero, with the exception of the pressure gradient, which is assumed to be constant. And, the volumetric flow rate of polymer solution and bore fluid is specified.
2. Wall condition: As nonslip condition holds true at the wall boundary, normal and tangential velocity components are zero.

$$v_n = v_s = 0 \quad (7)$$

3. Free surface condition: In our simulation, the shape of the extrudate is not known in advance; a free surface is used to represent the outer surface of the extrudate. Its position is computed as part of the solution. We assume that inner and ambient pressures are balanced, and so the normal force imposed on the free surface is zero.

$$f = 0 \quad (8)$$

Since no mass crosses the free surface, then

$$v \cdot n = 0 \quad (9)$$

where v is the velocity vector and n is the normal vector to the free surface.

4. Moving interface condition: A moving interface is similar to a free surface, in that its position is computed as part of the solution. At the interface, velocity vector and surface-force vector is continuous, so

$$v_1 = v_2 \quad (10)$$

And

$$f_1 = f_2 \quad (11)$$

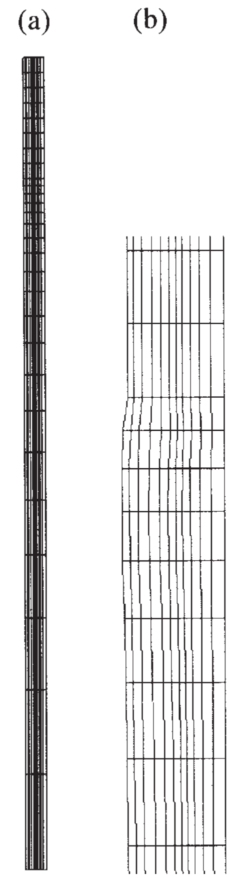

Figure 2 (a) initial mesh (b) final mesh enlarged near the die exit.

As we previously assumed that solvent and nonsolvent mass transfer is blocked by the thin layer, so

$$v \cdot n = 0 \quad (12)$$

That is, no mass across the interface.

If continuity equation and momentum conservation equations, these coupled ones, are solved simultaneously provided that we have a constitutive equation relating the stress with the rate of deformation, we can get the diameter and velocity profiles of the hollow fibers in the air gap. The two-dimensional flow problem is solved using finite elements method. The flow domain is discretized by means of quadrilateral elements. The interpolation methods for domain coordination and pressure are linear and quadratic for velocity. Because the free surface and moving interface's position is unknown, the flow domain is remeshed during the calculation, and the boundary nodes' moving direction is the normal to the initial mesh. In Figure 2, the initial mesh and the die swell part near the die exit are shown.

5. Outlet condition: If end velocity and tensile force are not specified, the outlet condition for this simulation is zero normal force and zero

TABLE I
The Parameters Used in the Simulation

Simulation parameters	Set value
Spinneret length	3 mm
Air-gap length	17 mm
Bore fluid density	1 g/cm^3
Bore fluid viscosity	1×10^{-3}Pa s
Dope density	1 g/cm^3
Dope viscosity	50 Pa s
Bore fluid rate	0.004 cm^3/s
Dope fluid rate	0.015 cm^3/s

tangential velocity, which replaces a long channel at the exit of the flow domain by a single boundary condition. It means though in this simulation the air-gap length is only 17 mm, the downstream flow behavior can be properly predicted.

RESULTS AND DISCUSSION

In Table I, the parameters generally used in the simulation are summarized (if parameters are not specified in later figures they are the same as ones showed in Table I). These parameters are based on the experimental data and rounded. In Figures 3 and 4, the typical velocity profiles, both on the free surface and on the interface of the hollow fiber, are presented. It can be seen that except at the position that is very near the spinneret the *r* component of the velocity vector is almost zero. That means, if no die swell is considered, one-dimensional simulation is acceptable. The enlarged swell part of the hollow fiber is shown in Figure 5. Therefore, at the exit of the spinneret the dope flow (left one) pushes the bore fluid down, which makes the inner diameter smallest at this position.

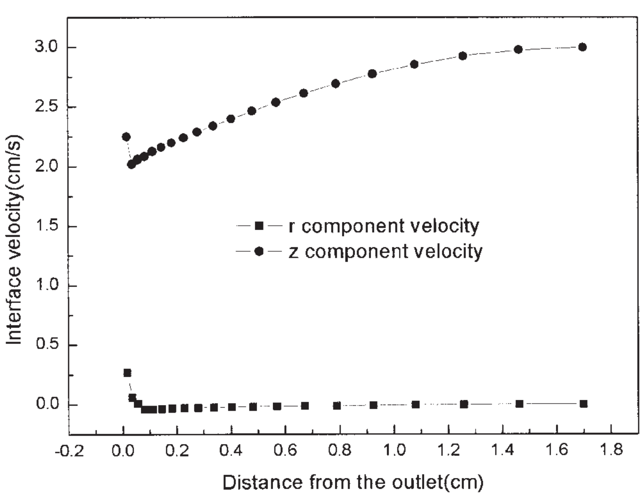

Figure 3 The velocity profile at the interface of the hollow fiber.

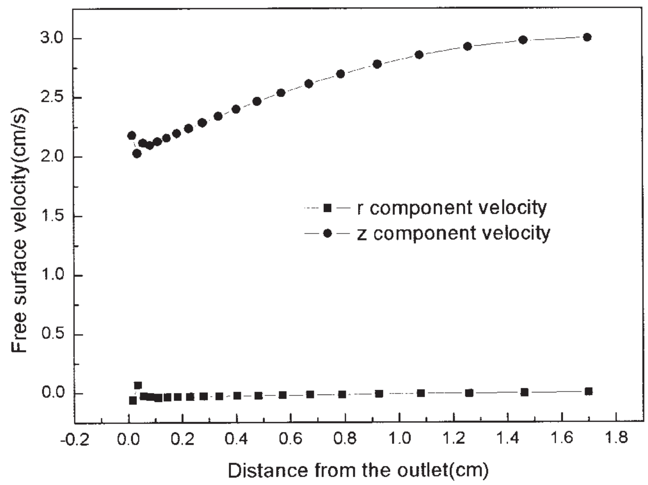

Figure 4 The velocity profile at the free surface of the hollow fiber.

Effect of inertia term on diameter profile

In some simulations, inertia term is always neglected.[5] Figure 6 shows the outer and inner diameters of the hollow fiber to illustrate the effect of the inertia term. As easily seen in Figure 6, inertia term almost has no effect on this simulation. The reason for the phenomena is that, in this procedure, spinning speed is relatively low (2.5 m/s). That is also the reason for the neglect of air friction.

Effect of gravity term on diameter profile

In some one- or two-dimensional simulations, gravity term is also neglected. However, it is known from the

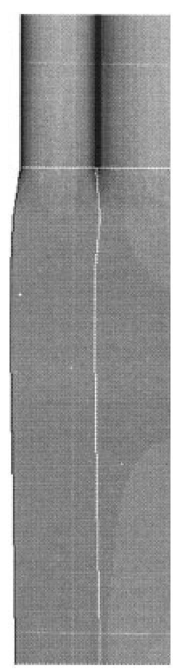

Figure 5 Enlarged die swell.

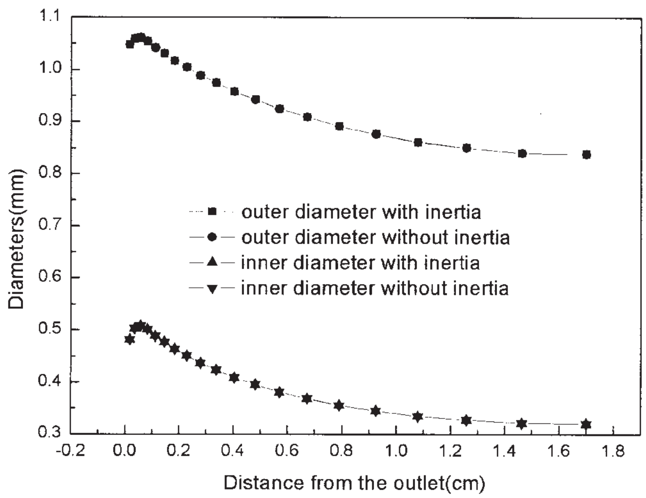

Figure 6 The effect of inertia term on the diameter profile of hollow fibers.

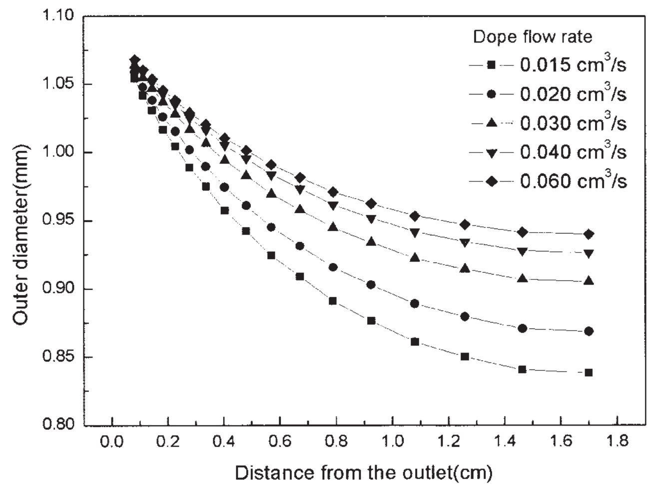

Figure 8 The influence of dope flow rate on the outer diameter of the hollow fiber.

experimental observation that gravity will push the hollow fiber toward the axis of symmetry and decrease both the inner and outer diameter. Figure 7 proves this phenomenon. As gravity term added to the momentum equation, the diameters of the hollow fiber decrease rapidly in the air gap compared with no gravity added one.

Effect of dope flow rate and bore fluid rate on diameter profile and thickness

On the basis of the experimental observation, the outer diameter will increase as dope flow rate increases. Similarly, the outer diameter will increase too as the bore flow rate increases. This is also approved is the simulation as showed in Figures 8 and 9. The reason is clear and easy to understand. As the bore flow rate increases, the inner diameter of the hollow fiber will increase. But, the result is not as simple as the dope flow rate increases. At small dope flow rate, Figure 10 illustrates that an increase of the dope flow rate will cause the inner diameter to increase. But, at large dope flow rate it decreases. It may be explained that at low flow rate, the effect are over neutralized by the die swell near the spinneret, as seen in Figure 11, that at small flow rate the die swell effect increases rapidly with the increase of dope flow rate, but at large flow rate it slow down. On the other hand, at quite large flow rate (0.06 cm^3/s), the effect cannot be neutralized completely and it will affect the inner diameter contrarily. It will push the bore fluid down and cause small inner diameter. Figure 12 shows that as dope flow rate increases the hollow fiber's thickness will increase too.

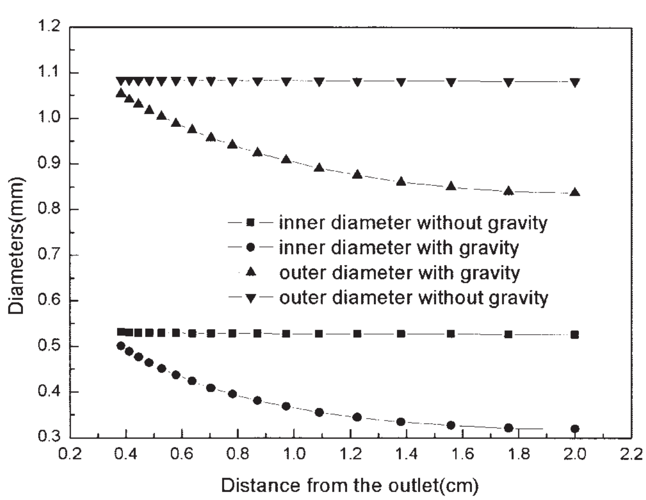

Figure 7 The effect of gravity term on the hollow fiber diameter profile.

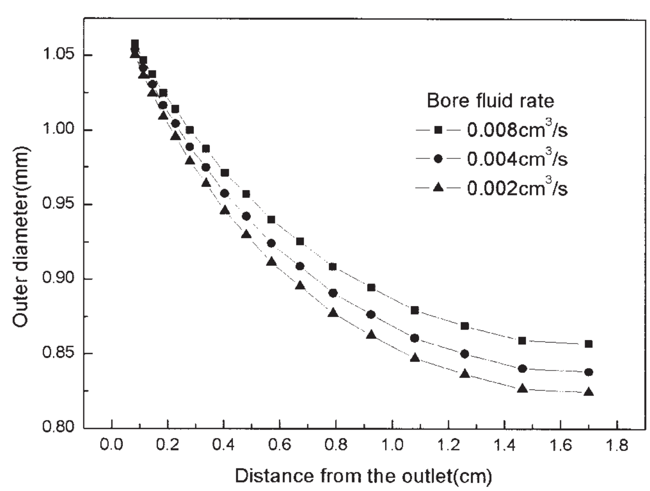

Figure 9 The influence of bore fluid rate on the outer diameter of the hollow fiber.

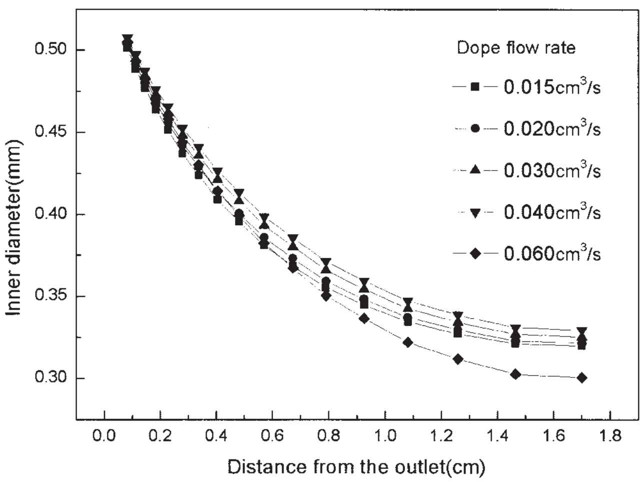

Figure 10 The influence of dope flow rate on the inner diameter of the hollow fiber.

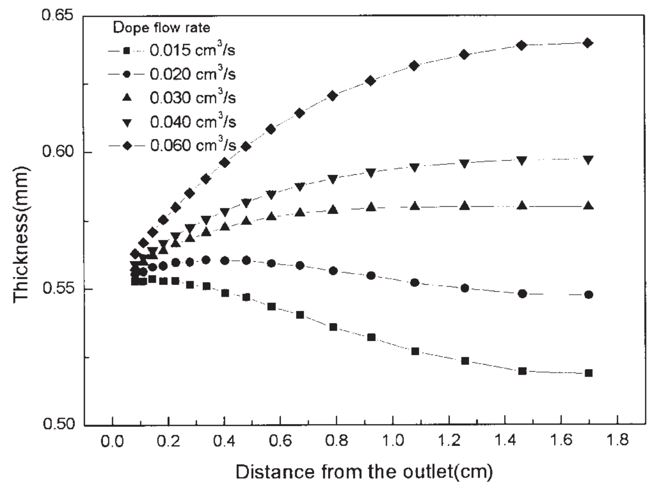

Figure 12 The influence of dope flow rate on the thickness of the hollow fiber.

Effect of dope viscosity on diameter profile

Dope viscosity plays an important role in controlling the diameters of the hollow fiber. Figures 13 and 14 show that the inner and outer diameters are easier to change along the distance as the dope solution's viscosity decreases. The inherent of this phenomena is that high viscosity fluid is unwilling to change under outer forces (here gravity act as this role). If high viscosity dope solution is used, but thin diameter is desired, the way to control is represented later in this article as imposing tensile force or control the end velocity of outlet.

Effect of tensile force on diameter profile

In high-speed melt-spinning, tensile force cannot be neglected, because it accelerates the fiber in the axial direction, forming a thin filament. But in hollow fiber-spinning, tensile force is not so important because usually no force will be imposed on it. Even so, the influence of tensile force on the formation of hollow fiber is investigated. The result is reasonable that the larger force imposed on it, the larger deformation it shows in the air gap. As the tensile force is relatively large (in this case 4×10^{-4}N), the extruded hollow fibers become unsmooth, as seen in Figure 15. If tensile force is larger than 4.5×10^{-4}N, the calculation cannot converge. This means the hollow fiber will break in such a large force.

Effect of end velocity on diameter profile

The influence of end velocity is very similar as tensile force that larger end velocity imposed on it, the larger

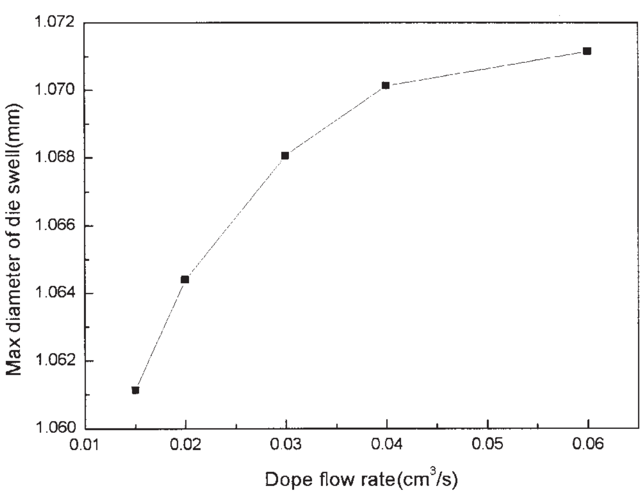

Figure 11 The effect of dope flow rate on the die swell phenomena.

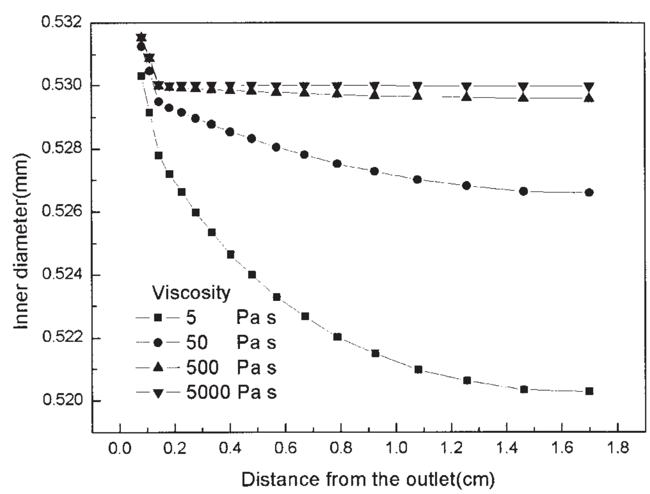

Figure 13 The effect of viscosity parameter on the inner diameter profile.

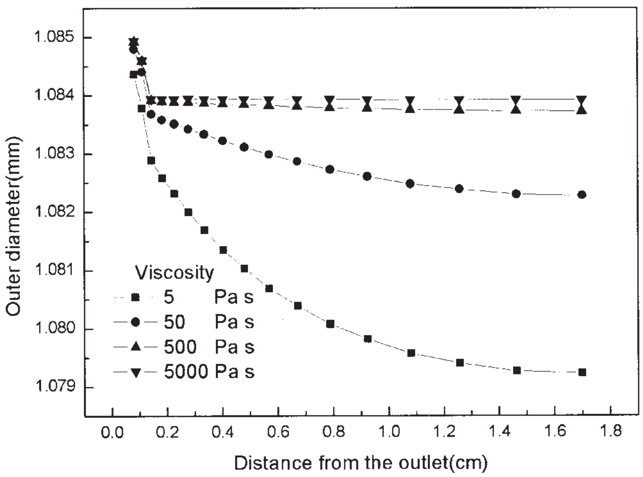

Figure 14 The effect of viscosity parameter on the outer diameter profile.

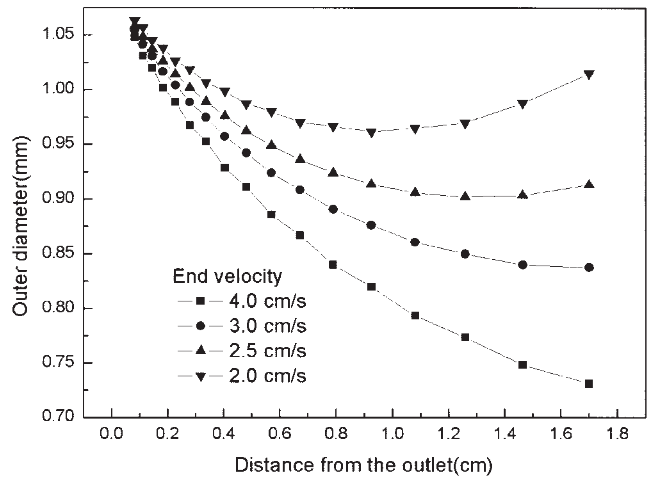

Figure 16 The effect of end velocity on the outer diameter of the hollow fiber.

deformation it shows in the air gap. But in real hollow fiber-spinning operation, end velocity is easier to be controlled and tested than tensile force. As shown in Figure 16, the outer diameter of hollow fiber will first become small then large again if end velocity is low. This phenomenon can only occur in the simulation, while in real world the fiber will not become large again, instead it will distort.

Effect of non-newtonian dope solution on diameter profile

For simplification, the dope solution previously discussed is all Newtonian. But it is not true for most dope solutions at high strain rate. Their viscosity is a function of the shear rate. Here, the simplest empiricism expression power law is investigated to check the influence of non-Newtonian to the formation of the hollow fibers. On the basis of eq. (3), m is given as 70 and 50 (Pa s^n) while n is given as 0.5081.[15] Power law states that the fluid's viscosity will decrease when shear rate is increased. Combined with viscosity influence in 3.4, it is easy to understand the phenomena, showed in Figure 17, that at same initial viscosity power law fluid is easier to deform along the distance.

CONCLUSIONS

The deformation of hollow fiber in the air gap (close to the spinet) is numerically simulated using the finite element method. On the basis of the simulated results, inertia term can safely be neglected in low-speed hollow fiber-spinning, but the influence of gravity cannot be neglected. Some common sense is proved: increasing the volume rate of dope and bore flow will in-

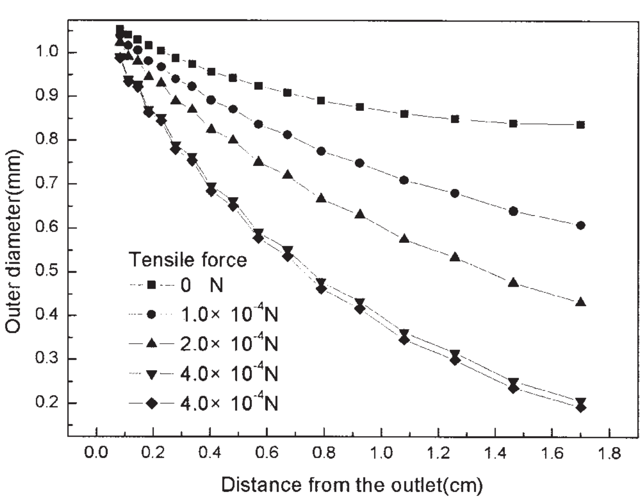

Figure 15 The effect of tensile force on the outer diameter of the hollow fiber(unsmooth phenomena).

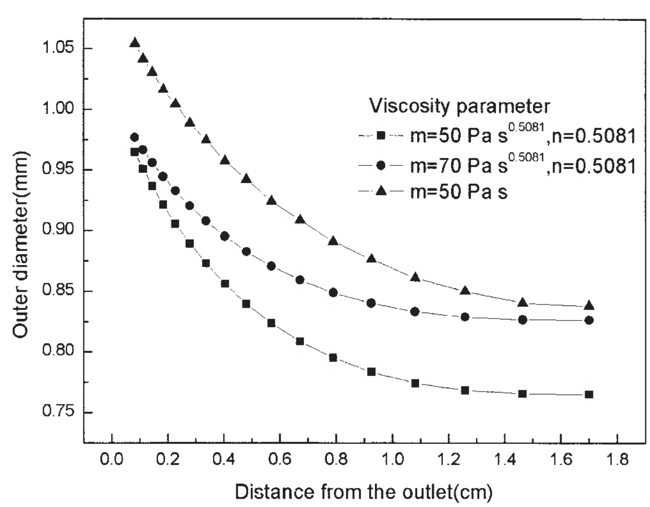

Figure 17 The effect of non-Newtonian character on the outer diameter of the hollow fiber.

crease the outer diameter of the fibers; large tensile force or large end velocity will cause large deformation in the air gap. And, larger viscous dope solution tends to make less deformation in the air gap. It is also found that an increase of the dope flow rate at small dope flow rate resulted in an increase of the inner diameter, while at large dope flow rate it decreased. Although many effects have been discussed here, two important elements, surface tension and elastic effect, are not investigated. They are valuable to investigate in later work.

NOMENCLATURE

Symbols

m parameter of power law
n parameter of power law
p pressure(Pa)
g gravitational function (980 cm s^{-2})
f_i forces (N)

Greek

vr velocity of *r*-component of the hollow fiber(cm s^{-1})
v_z velocity of *r*-component of the hollow fiber(cm s^{-1})
η viscosity(Pa s)
$\dot{\gamma}$ magnitude of the rate-of-strain tensor
ρ_h density of hollow fiber (g cm^{-3})
τ_{ij} shear stress (N cm^{-2})
τ_{ij} normal stress (N cm^{-2})
v_n normal component of velocity vector(cm s^{-1})
v_s tangential component of velocity vector(cm s^{-1})

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