Modeling of fiber spinning flows of pre-ceramic polymer melts

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Abstract

Pre-ceramic polymers are thermally decomposed in a controlled atmosphere into non-oxide covalently bonded ceramics.A melt spinning process is used to prepare ceramic fibers followed by the thermal conversion of green fibers.The extensional viscocity of polymer-derived ceramics (PDCs) can be computed based on the measurement of fiber diameter at the exit from the die. The final goal of this study is to establish correlations between the rheological properties in shear and extension for PDCs, in order to determine the proper working condition for obtaining high quality fibers.

1 Introduction

Non-oxide ceramic fibers have various useful properties such as thermal shock resistance,creep resistance and relatively high tensile strength and modulus values.Thermally decomposing preceramic polymers for the preparation of fine diameter fibers showed great advantages.It offers good control of compositions and structures, and the ability to process particular shapes.
Significant progress has been made in the development of synthesis routes to pre-ceramic polymers ever since the first synthesis of SiC ceramic material from polycarbosilanes precursors.The silicon-based polymers have proven to be promising tractable compounds for production of technologically important ceramic components such as fibers, microfibers, protective coatings, porous materials or complex-shaped bulk parts.

One of the demanding problems is to develop polymers with appropriate rheology and melt stability to allow for continuous melt spinning.The melt-spinning process involves an extrusion process of the polymer from molten state and an on-line fiber drawing that stabilizes the fiber line, reduces the fiber diameter and alignment of the polymeric chains along the fiber-axis. Control of spinning and pyrolysis processes is required for the preparation of polymer derived ceramics with reliable and improved properties.

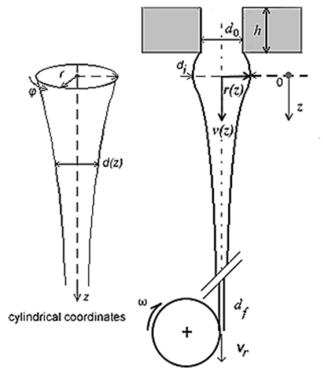

Fig. 1. Schematic representation of the fiber-spinning process.

Figure 1: Schematic representation of the fiber-spinning process.

A one dimensional model of an axial-symmetric fiber spinning process is used for analysis, with the following assumptions:
1.The melted polymer is considered to be incompressible fluid and to flow is steady.
2.Exit “elastic phenomenon” such as die swell is ignored. The position of 0=z is taken at the point of maximum die swell
3.For 0 $\geq$z the filament is assumed axisymmetric and that is does not rotate. The diameter of the fiber d(z) = 2R(z) is a monotonous decreasing function and the motion within the filament is considered pure-extensional. Velocity and normal stresses distributions are assumed to be uniform across the flow area
4.For steady and isochoric motions of thin fibers (d $<$ 1 mm), inertia, gravity, surface tension and air friction can be neglected.
Mass,momentum and energy balance yield the following equations:

$$Q = A(z) * v(z) = A_0 * v_0 = ct$$

$$0 = \frac{d}{dz}[A(z)(\sigma_{zz} - \sigma_{rr})]$$

$$\rho C_p v(z)\frac{dT}{dz} = \frac{-2h}{R(z)}(T(z) - T_0) + (\sigma_{zz} - \sigma_{rr})\frac{dv}{dz}$$

where Q is the flow rate, $v(z)$ and $A(z) = \pi R^2(z)$ are the velocity and the cross-section area of the fiber.$\sigma_{zz} - \sigma_{rr}$ is the tensile strength within the fiber (σzz and σrr are the normal stresses on the z-direction and r-direction of the fiber,respectively). ρ is the mass density ; C_p is the heat capacity , h is the heat transfer coefficient at the surface of the filament , $T(z)$ is the polymer temperature along the spin line , T_0 is the temperature of the environment. $\epsilon = \frac{dv}{dz}$ is defined as the characteristic extensional strain rate of the spinning motion.
It is essential to recognize that fiber spinning process involve extensional deformation and the extensional viscosity is the unique material function to describe its rheology. It is expressed as:

$$\eta_e(\epsilon, T) = \eta_e(\epsilon, T)[\frac{E}{R}(\frac{1}{T} - \frac{1}{T_0})]$$

where E is the activation energy for this material and R is the ideal gas constant.
An exponential or polynomial function is used to approximate the diameter of the fiber along the spinline, which is further used in a non-dimensional form to determine the melt extensional viscocity.

2 Experimental

The equations were solved for the following values:

$$Q = 64.44 \;\; mm^3/min$$
$$v_r = 117.34 \;\; m/min$$
$$T_s = 117^o \;\; C$$
$$d_0 = 0.2 \;\; mm$$
$$T_0 = 25^o \;\; C$$
$$z = [0\; 0.5] \;\; mm$$

The solution was obtained for two cases:
1.Approximating radius of the fiber along the spinline with a linear function and solving for η,dv/dz and r.
2.Taking viscocity as constant and solving for A,v,d/dz and T.

RK4 technique,along with the shooting method, was used to solve the differential equations using $ode45$ function in MATLAB.For the second case, the equations were solved taking a draw ratio of 10,20 and 30.

3 Results

The following graphs were obtained :

3.1 Case 1

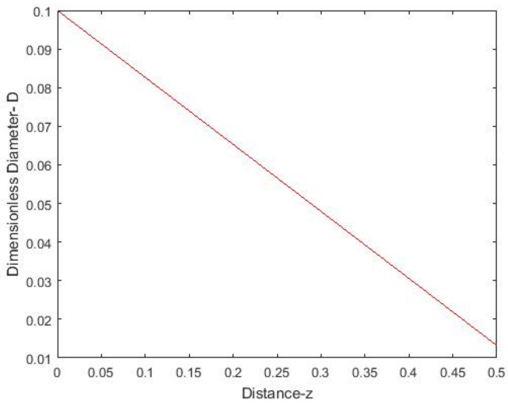

Figure 2: Varitaion of dimensionless diameter, $D = d(z)/d_0$ with z.

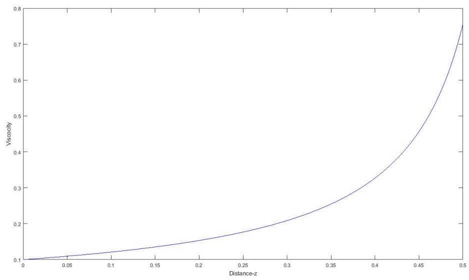

Figure 3: Varitaion of η with z.

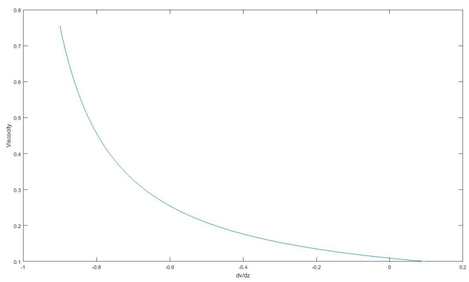

Figure 4: Varitaion of η with $\frac{dv}{dz}$.

3.2 Case 2

3.2.1 Draw Ratio=10

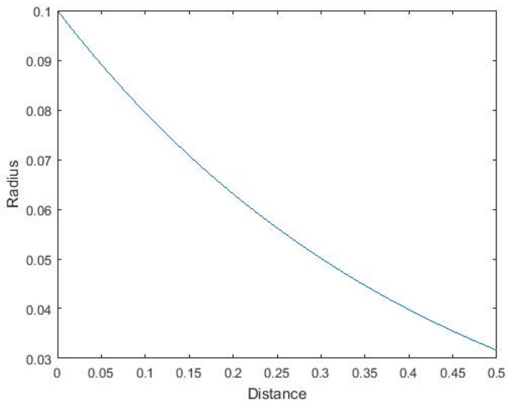

Figure 5: Varitaion of $R(z)$ with z.

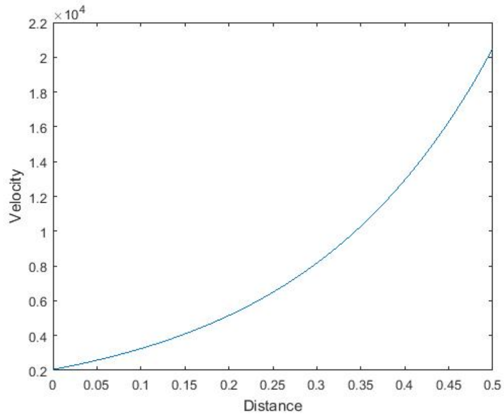

Figure 6: Varitaion of $v(z)$ with z.

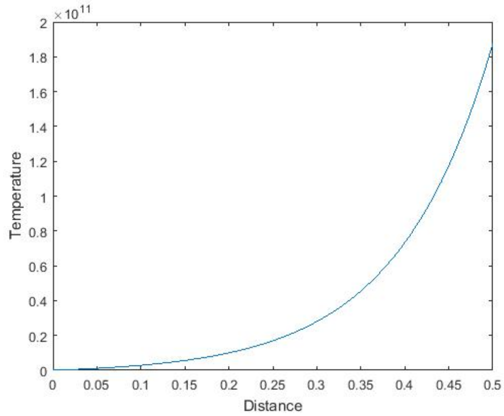

Figure 7: Varitaion of $T(z)$ with z.

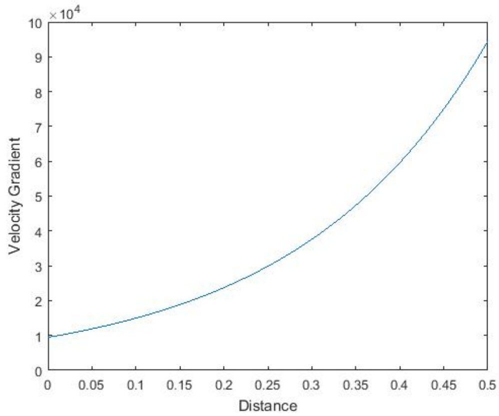

Figure 8: Varitaion of $\frac{dv}{dz}$ with z.

3.2.2 Draw Ratio=20

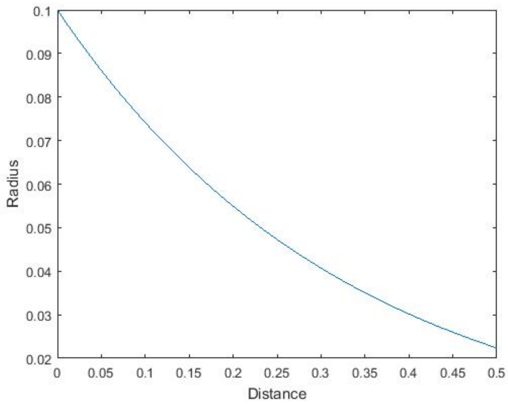

Figure 9: Varitaion of $R(z)$ with z.

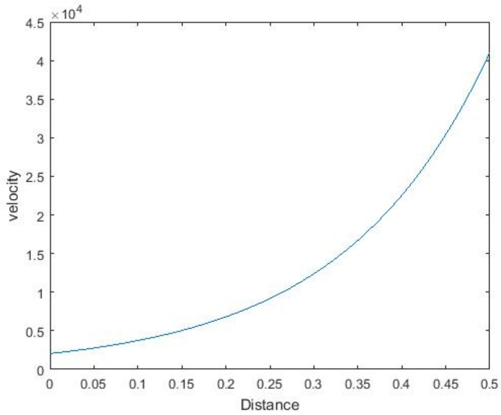

Figure 10: Varitaion of $v(z)$ with z.

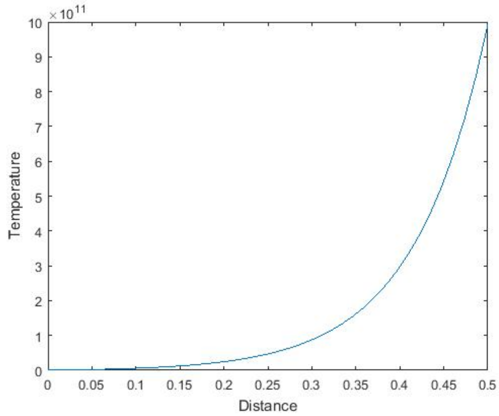

Figure 11: Varitaion of $T(z)$ with z.

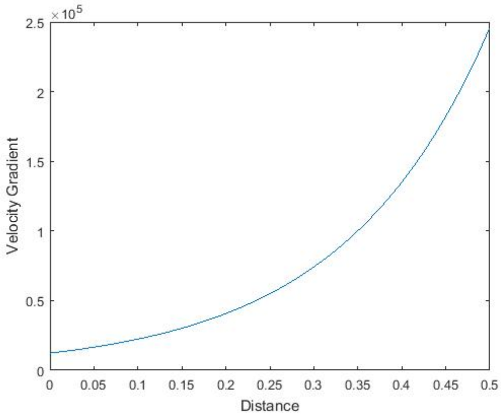

Figure 12: Varitaion of $\frac{dv}{dz}$ with z.

3.2.3 Draw Ratio=30

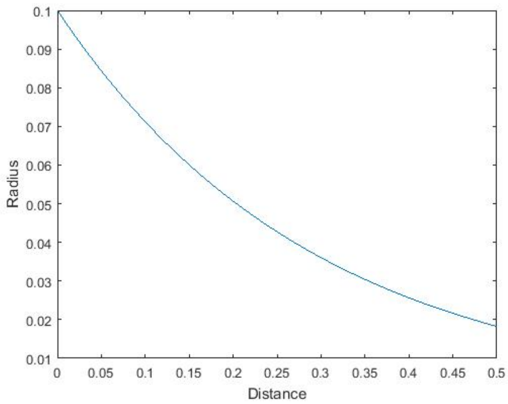

Figure 13: Varitaion of $R(z)$ with z.

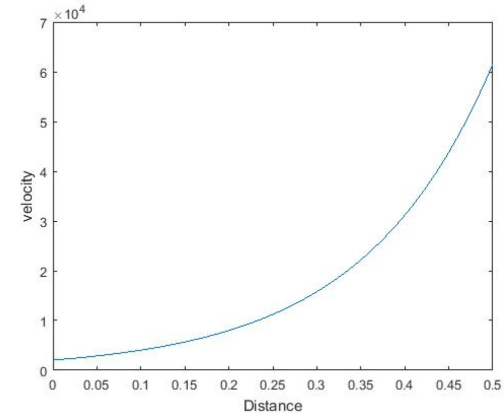

Figure 14: Varitaion of $v(z)$ with z.

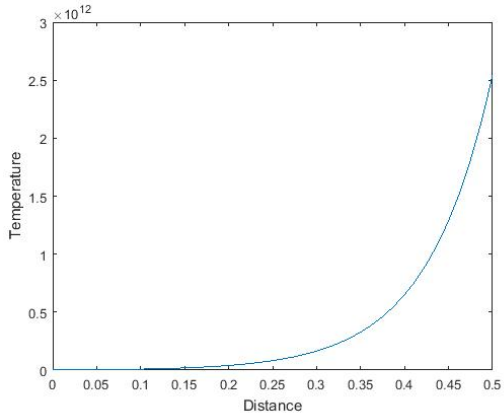

Figure 15: Varitaion of $T(z)$ with z.

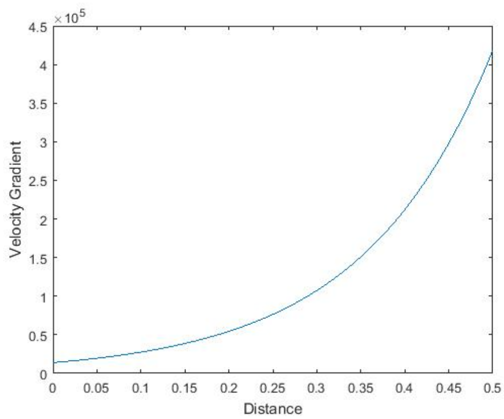

Figure 16: Varitaion of $\frac{dv}{dz}$ with z.

4 Conclusion

Modeling of PCS pre-ceramic polymer in fiber spinning process was performed in this work, based on the measurements of fiber diameter along spinline during the process.
The equations obtained by mass,momentum and energy balance were solved using RK4 technique combined with the shooting method.