

# The polymer melt swelling behavior in micro-extrusion process for preparation of continuous carbon fiber reinforced PP prepreg filament

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**Abstract.** The micro-extrusion process for polymer melt has been increasingly used for the production of high value-added prepreg filaments with applications in the fields of fused deposition modeling (FDM). In this study, the effect of characteristic scale, defined as the gap of die land in an annular micro-extrusion die, on the extrudate swelling behaviour of viscoelastic melt is analyzed through numerical simulations and micro-extrusion experiments. The results show that the swelling behaviour displays an obvious dependence on the characteristic scale. An increase in the characteristic scale reduces the swell ratio and retards the process to reach the equilibrium state. In contrast, a decrease in the characteristic scale results in a larger magnitude of change in velocity field and faster relaxation development of stress field. The location of the maximum velocity layer for the laminar flow gradually deviates from the geometric center of channel toward the wall of mandrel with the increase in the characteristic scale. It is imperative to consider the complicated swelling behavior and remarkable viscoelastic effect simultaneously in micro-extrusion process.

## 1 Introduction

The micro-extrusion process for polymer melt is one of the most important micromolding technologies<sup>[1, 2]</sup>, and has been increasingly used for the production of high value-added prepreg filaments with applications in the fields of fused deposition modelling (FDM)<sup>[3]</sup>, such as fused filament fabrication (FFF) and fused granulate fabrication (FGF). Maarten and Lucas<sup>[4]</sup> proposed an apparatus where the matrix material, preferably thermoplastic, is fed in a screw-type extruder in a pellet form and through a channel that encloses the fibre feed system and impregnates the introduced fibre tow. It is suggested that the fibre tow is previously impregnated with the polymer matrix material to enhance adhesion. However, the previously unknown swelling behaviour emerging from the micro-extrusion die can easily lead to a complicated extrudate deformation of the shape and dimension of the prepreg filaments<sup>[5]</sup>. Owing to the distinguishing feature of the flow channel with the characteristic scale at the micron level, the effect of extrudate swell behaviour on the accuracy and precision of the prepreg filaments becomes much more significant than that in the conventional

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extrusion process<sup>[6, 7]</sup>. Here, the term “characteristic scale” is adopted to represent the channel size in different types of the micro-extrusion die due to the flexibility and extensibility of the meanings, such as the diameter  $D$  in the capillary die<sup>[8]</sup>, the height  $H$  in the slit die, the gap  $H$  in the annular die, and the average gap thickness  $D_C$  in the micro profile die. Therefore, it is crucial to investigate the extrudate swell behavior given the micro-extrusion die design and process control.

Therefore, the primary purpose of this study is to investigate the extrudate swelling behaviour of viscoelastic polymer melt in the micro-extrusion process for preparation of continuous carbon fibre reinforced polypropylene (PP) prepreg filament. The double convected pom-pom (DCPP) constitutive model is employed to describe the viscoelastic rheological behaviour of a polypropylene melt flowing through an annular micro-extrusion die. A full three-dimensional (3D) numerical simulation with different characteristic scales of annular channels was conducted to analyse the effect of the characteristic scale on the extrudate swelling behaviour. The extrudate swelling ratios of the inner and outer diameters were used to characterize the evolution behaviour. The distribution of the velocity field is discussed to explain the discrepancy in the swelling behaviour in different flow channels. Finally, The micro-extrusion experiments were performed to validate the model behaviour and confirm its predictive power for the swelling behaviour.

## 2 Numerical modeling

### 2.1 Simulation design and method

#### 2.1.1 Governing equations

Considering the characteristics of the micro-extrusion process when the stable extrusion arrives, the assumption of isothermal steady laminar flow is also made. The influences of the inertial force, surface tension, and gravity effect are ignored. The governing equations including the continuity and momentum balance are derived as follows in tensor notation:

$$\nabla \cdot \mathbf{v} = 0 \quad (1)$$

$$-\nabla p + \nabla \cdot \boldsymbol{\tau} = 0 \quad (2)$$

where  $\nabla$  represents the Hamilton differential operator,  $\mathbf{v}$  is the velocity vector, and  $p$  denotes the hydrostatic pressure.  $\boldsymbol{\tau}$  is defined as the total extra stress tensor, which is split into the sum of two components, namely the purely viscous component  $\boldsymbol{\tau}^v$  and the viscoelastic extra stress component  $\boldsymbol{\tau}^E$ , where  $\boldsymbol{\tau}^v = 2\eta_2 \mathbf{D}$  and  $\eta_2$  denote the Newtonian viscosity. The total extra stress tensor,  $\boldsymbol{\tau}$ , for the viscoelastic melt can be computed as follows:

$$\boldsymbol{\tau} = 2\eta_2 \mathbf{D} + \boldsymbol{\tau}^E. \quad (3)$$

Therefore, a constitutive equation that associates  $\boldsymbol{\tau}^E$  with the velocity or velocity gradients is necessary to complete the set of governing equations. In the present work, we applied the differential DCPP model for the viscoelastic flow and extrudate swell simulation, which can provide qualitative information on the macromolecular behavior. The DCPP model calculates  $\boldsymbol{\tau}^E$  based on the following algebraic formulation with corrective factor  $(1-\zeta)$ :

$$\boldsymbol{\tau}^E = \frac{G}{1-\zeta} (3\mathcal{A}^2 \mathbf{S} - \mathbf{I}), \quad (4)$$

where  $G$  denotes the shear modulus of the melt and  $\zeta$  is the nonlinear material parameter that controls the ratio of the second to first normal stress differences.  $\mathbf{S}$  and  $\mathcal{A}$  are the state variables of the macromolecular, which represent the orientation tensor and stretching scalar,

respectively, and  $\mathbf{I}$  is defined as the unit tensor. The two state variables,  $\mathbf{S}$  and  $A$ , are calculated as follows:

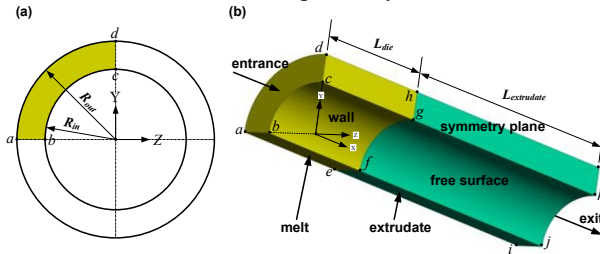
$$\lambda \left[ \left(1 - \frac{\xi}{2}\right) \overset{\nabla}{\mathbf{S}} + \frac{\xi}{2} \overset{\Delta}{\mathbf{S}} \right] + \lambda(1 - \xi) [2\mathbf{D} : \mathbf{S}] \mathbf{S} + \frac{1}{A^2} \left[ \mathbf{S} - \frac{\mathbf{I}}{3} \right] = 0, \quad (5)$$

$$\lambda_s \frac{DA}{Dt} - \lambda_s (\mathbf{D} : \mathbf{S}) A + (A - 1) e^{\frac{2(A-1)}{q}} = 0, \quad (6)$$

where  $\mathbf{D}$  is defined as the rate of deformation tensor and is calculated as  $\mathbf{D} = [(\nabla \mathbf{v}) + (\nabla \mathbf{v})^T]/2$ ; here,  $(\nabla \mathbf{v})$  denotes the velocity gradient and  $(\nabla \mathbf{v})^T$  is the transpose of the velocity gradient. The two superscripts  $\nabla$  and  $\Delta$  of  $\mathbf{S}$  correspond to the upper- and lower-convected derivatives, respectively.  $\lambda$  and  $\lambda_s$  are the melt relaxation times associated with the orientation and stretching mechanisms of macromolecules, respectively, and  $q$  characterizes the number of dangling branches connected at both extremities of the pom-pom molecule.

### 2.1.2 General geometry and boundary conditions

The 3D flow domain of the melt flowing through an annular die at a given microlevel characteristic scale of  $H$  ( $H = R_{out} - R_{in}$ ) is depicted in Fig. 1. Owing to the symmetry of the annular die, only one-quarter of the flow domain shown in Fig. 1(a) was calculated for all simulations to obtain lower computational time. The calculated flow domain contains two subdomains with lengths of  $L_{die}$  and  $L_{extrudate}$ , as depicted in Fig. 1(b), which denote the melt flow regions inside and outside of the die, respectively.



**Fig. 1.** Flow domain of an annular die: (a) cross-section, (b) one quarter is considered for all simulations.

Five groups of flow channels with a characteristic scale of  $H$  ( $H = R_{out} - R_{in}$ ) ranging from 0.2 mm to 1.0 mm were considered, as shown in Table 1.

**Table 1.** Geometries of channels and corresponding flow rates

channel series	$R_{in}$ (mm)	$R_{out}$ (mm)	$H$ (mm)	$L_{die}$ (mm)	$L_{extrudate}$ (mm)	$S_{die}$ (mm <sup>2</sup> )	$Q$ (mm <sup>3</sup> /s)
(a)	1.5	1.7	0.2	5	10	2.011	4.021
(b)	1.5	1.9	0.4	5	10	4.273	8.545
(c)	1.5	2.1	0.6	5	10	6.786	13.572
(d)	1.5	2.3	0.8	5	10	9.550	19.101
(e)	1.5	2.5	1.0	5	10	12.566	25.133

### 2.1.3 Quantification for the swell behavior

Fig. 1(a) displays the schematic of the cross-section of an annular channel. Two swell ratios, i.e.,  $B_I$  and  $B_O$ , are defined as in equation (7) to provide a quantitative analysis of the different possible extrudate swell behaviors in the micro-extrusion process. They denote the swelling behavior in the extrudate inner diameter and outer diameter, respectively.

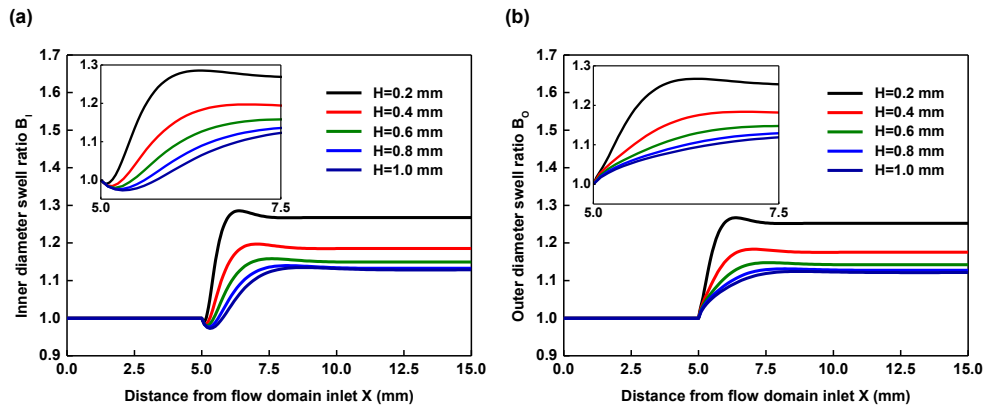
$$B_I = \frac{R_{in-extrudate}}{R_{in-die}}, B_O = \frac{R_{out-extrudate}}{R_{out-die}} \quad (7)$$

where  $R_{in-die}$  and  $R_{out-die}$  represent the inner and outer diameters of the annular die,  $R_{in-extrudate}$  and  $R_{out-extrudate}$  denote the inner and outer diameters of the extrudate, respectively.

## 3 Results and discussion about extrudate swelling behavior

### 3.1 The effect of characteristic scale in micro-extrusion

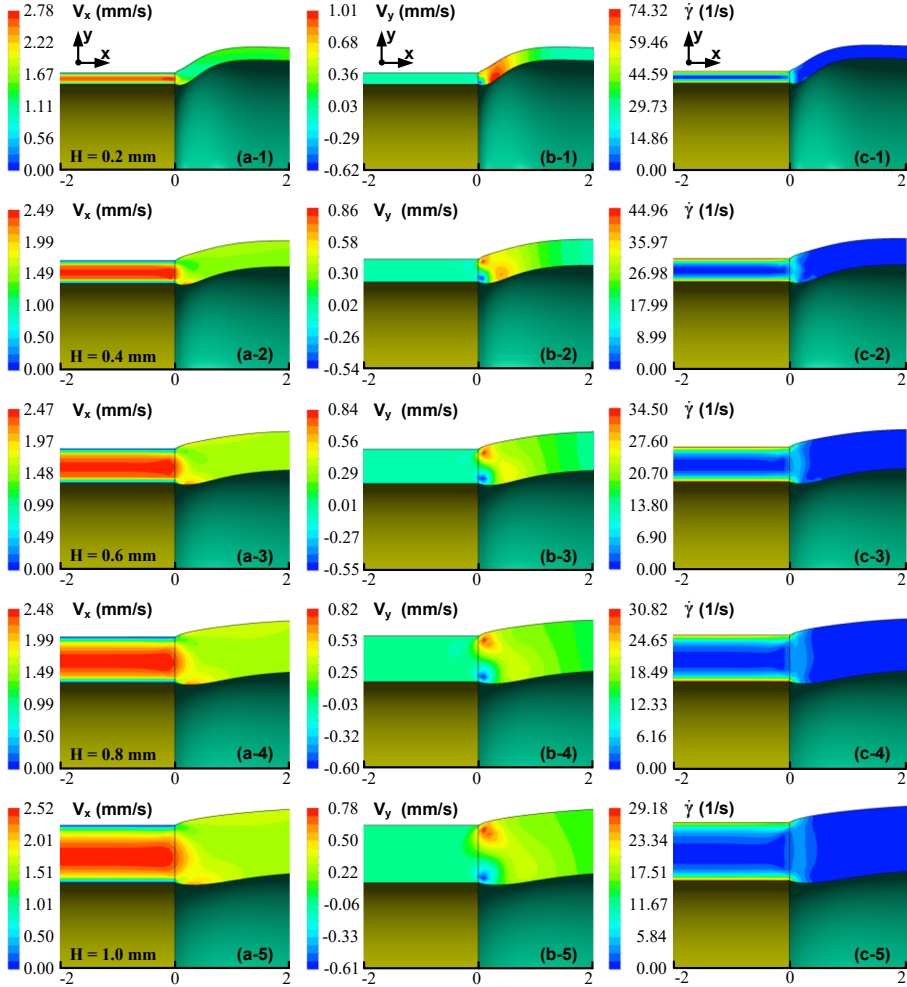
The predicted variations of the two important swelling ratios defined in equation (7) are plotted in Figs. 2(a)–(b). Fig. 2(a) shows the comparison of the inner diameter swell ratio,  $B_I$ , at different characteristic scales with the typical characteristic of continuous evolution from the die exit. The inner diameter swell ratio decreases instantaneously with respect to the negative extrudate swell ratio ( $B_I < 1$ ) upon leaving the die exit for a distance of 1 mm. Then, it increases rapidly to its maximum value within a short distance. Immediately thereafter,  $B_I$  progressively declines to reach an equilibrium value. Fig. 2(b) shows the comparison of the outer diameter swell ratio,  $B_O$ , at different characteristic scales. In contrast with the swell behavior of the inner profile near the die exit, the outer profile does not display the so-called negative extrudate swell. It increases directly and quickly after leaving the die exit. Similarly, after increasing monotonously to a maximum value, the outer diameter swell ratio decreases gradually to an equilibrium value.



**Fig. 2.** Dependence of extrudate swell ratio predicted using the viscoelastic model on the characteristic scale in micro-extrusion at the same average velocity  $v$  of 2 mm/s: (a) inner diameter swell ratio  $B_I$ , (b) outer diameter swell ratio  $B_O$ .

In Fig. 3(a), the flow velocity inside the die shows a concentric distribution, and the flow is relatively faster along the flow channel centerline. For the lowest value of  $H = 0.2$  mm, the maximum velocity component,  $v_x$ , is larger than that for a higher value of  $H$ . Moreover, the melt layer with the maximum velocity gradually moves away from the wall of the die and approaches the wall of the mandrel with the increase in the value of  $H$ . This is an important

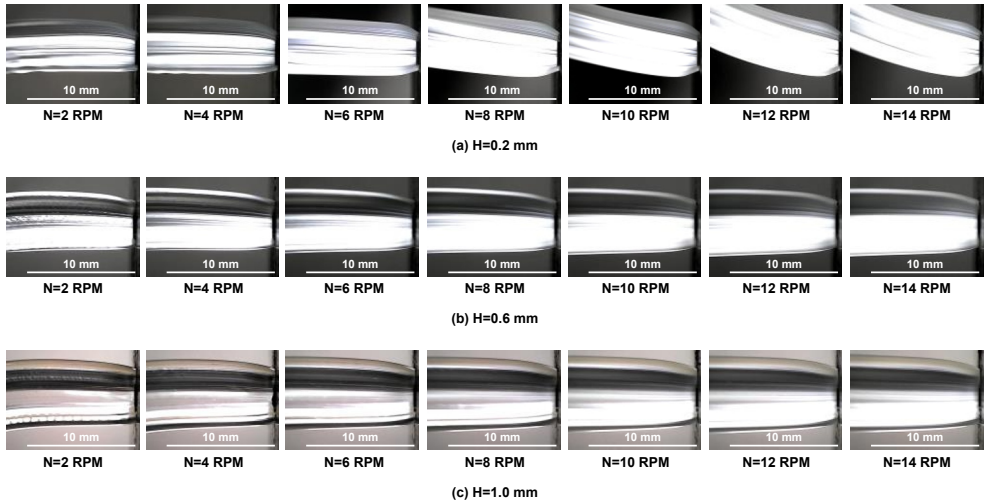
factor explaining why increasing the value of the characteristic scale leads to a smaller extrudate swell ratio,  $B_O$ , as shown in Fig. 2. Fig. 3(b) displays the effect of the characteristic scale on velocity component  $v_y$ , which is directly related to the radial extrudate swell behavior outside the die. The maximum value of  $v_y$  decreases with an increase in the characteristic scale. On the contrary, an increase in the value of the characteristic scale makes the flow more uniform owing to the smaller local shear rate, as shown in Fig. 3(c).



**Fig. 3.** Contours of field variables of melt flowing through different channels at the same average velocity  $v$  of 2 mm/s: (a) axial velocity,  $v_x$  (b) radial velocity,  $v_y$  and (c) local shear rate,  $\dot{\gamma}$ .

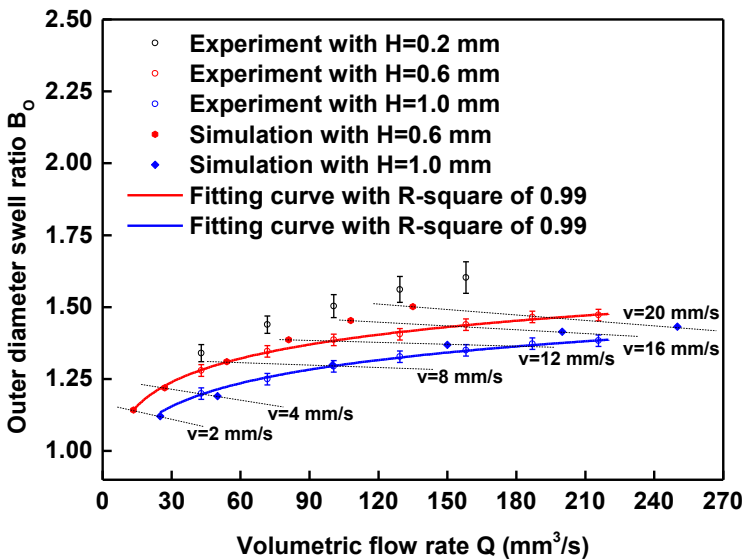
### 3.2 Experiment verification of the simulation results

Fig. 4 displays the outer profile of extrudate along the flow direction in the micro-extrusion experiment with the screw speed ranging from 2 RPM to 14 RPM. It can be observed that there exists an obvious increase in diameter of the outer profile near the die exit, as shown in Figs. 4(a), 4(b), and 4(c), respectively.



**Fig. 4.** The outer profile of extrudate along the flow direction in the micro-extrusion experiment with the screw speed ranging from 2 RPM to 14 RPM : (a)  $H=0.2\text{mm}$ , (b)  $H=0.6\text{mm}$ , (c)  $H=1.0\text{mm}$ .

Fig. 5 displays the predicted swell ratio  $B_o$  of the outer profile at different volumetric flow rates in comparison with the measurement results in the micro-extrusion experiment. In particular, the relationship between screw speed  $N$  and volumetric flow rate  $Q$  has been reported from the micro-extrusion experiment of PP melt using the HPE-100H extruder. When the volumetric flow rate is relatively small, the results from the numerical simulations are in good agreement with the experimental measurements, which proves the feasibility of the theoretical analysis and numerical simulations.



**Fig. 5.** The predicted swell ratio  $B_o$  of outer profile at different volumetric flow rates in comparison with the measurement results in the micro-extrusion experiment.

### 4 Conclusion

The extrudate swelling behavior exhibits an obvious dependence on the characteristic scale

of the channel in the micro-extrusion process. Increasing the characteristic scale  $H$  contributes to a smaller value of the extrudate swell ratio  $B$  and retards the swelling process of reaching the equilibrium state. The distributions of the velocity field at different characteristic scales,  $H$ , are investigated to elucidate the discrepancy of the swelling behavior. A larger magnitude of change in the velocity field and faster relaxation development for the stress field is caused by the decrease in the characteristic scale. The simulation results based on the DCP model are in good agreement with the experimental measurements at lower screw speed, which proves the feasibility of the theoretical analysis and numerical modeling.

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