

Numerical Simulation of the Non-Isothermal Co-Extrusion Fiber Spinning with Flow-Induced Crystallization

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ABSTRACT

In this work, the numerical simulation of the non-isothermal steady co-extrusion fiber spinning with flowinduced crystallization is explored. The model is based on the formulation originally proposed by China *et al.* in which Newtonian and Phan-Thien-Tanner (PTT) fluids are considered the core and the skin layer, respectively. The polymeric flow rate fraction, Deborah dimensionless number and the PTTs parameters on the temperature, the velocity and the crystallization profiles are analyzed. The numerical results show: the temperature profile is sensitive to the polymeric layer flow rate and the deformation parameters (shear thinning and extensional), the tensile stress induced crystallization parameter has a strong influence at the onset of the process, increasing drastically temperature and crystallinity.

Keywords: Polymer processing; PTT model; FIC.

NOMENCLATURE

A	cross sectional area	Tmax	maximum crystallization temperature
Ao	transversal section area at $z = 0$	T_a	environment temperature
σ	stress tensor	T_{0}	temperature at $z = 0$ of the fiber
σ_{rr}	stress in the radial direction	F	draw force applied at $z = L$
σzz	stress in the axial direction	ĸ	flow-induced crystallization factor
ϕ	the flow rate fraction of the skin-layer	п	avrami index
ر بر	shear thinning/thickening PTT parameter	а	dimensionless area
5	the automaional DTT nonemator	v	dimensionless velocity
ε	the extensional PTT parameter	τ_{ii}	dimensionless stress
Z	axial direction	v	dimensionless crystallinity parameter
D	strain tensor	λ 0	line single set and set and set
μ^{c}	core layer viscosity	θ	dimensionless temperature
5	skin laver viscosity	ζ	dimensionless axial position
μ		M	viscosity ratio
V O V	initial velocity	N	dimensionless first normal stress
V_L	final velocity		difference
E	elastic modulus	G	dimensionless axial force
λ	relaxation time	St	Stanton number
L	velocity gradient	De	Deborah number
$tr \sigma^s$	trace of stress tensor	Dr	draw ratio
ΔH_f	heat of crystallization per unit mass	ΔH_{f}^{*}	dimensionless crystallization heat
C_p	heat capacity of the skin fluid	J	
Ń	the crystallinity	$\Delta H_f X_{\infty}$	
ρ	skin fluid density	$\overline{C_nT_0}$	-
X_{∞}	maximum crystallinity	w	dimensionless axial strain

1. INTRODUCTION

Many economic and technical advantages have been found in co-extrusion, it has gained wide recognition a polymer processing technique achieving unique capable of product performance, by combining the properties of different materials at lower cost Hufenus et al. (2012), Perret et al.(2013), Reifler et al. (2014). Modeling and simulation of co-extrusion process require simultaneous consideration of constitutive and mass, momentum and energy conservation equations. The complete understanding of the process needs to take into ac-count important features reported in the literature like discontinuities at the interface, non-Newtonian behavior, encapsulation phenomena and the imple-mentation of numerical techniques (Liu and Beris 1988).

The fiber spinning theory has been studied by many research groups Kase and Matsuo (1965), Sung et al. (2005), Denn et al. (1975), Ziabicki (1976), Joo et al. (2002). Important aspects of stability and instability phenomenon observed in the spinning process have attracted researchers attention for both academic and industry Pearson and Matovich (1978), White and Ide (1978), Fisher and Denn (1976), Gelder (1971), Beris and Liu (1988). The transient solution of the fiber spinning with and without flow-induced crystallization (FIC) is indispensable to developing strategies for stabilization and optimization Lee and Park (1995). The FIC involves complex cinematics and thermal conditions, the phase transition is extremely sensitive to flow deformation and the material exhibits a non-Newtonian behavior. Consequently, a good under-standing of FIC dynamics is crucial in high-speed melt spinning modeling. Complete reviews of FIC can be found in McHugh (1990), Eder et al. (1990), Sung et al. (2005).

In regard of the interaction of two fluids, Lee and Park (1995) studied the draw resonance instability using a model where Newtonian and Upper-Convected Maxwell fluids (UCM) as the core and the skin layer, respectively. For an isothermal spinning flow, Ching-China and Jyh-Chau (1996) analyzed the influence of flow-rate ratio, Deborah number and deformation parameters (extensional and shear) on the spinline velocity. In their model Newtonian and PTT fluids were considered the core and the skin layer, respectively. For the non-isothermal coextrusion flow, Nordberg and Winter (1990) used the Warner model like the skin layer. They reported that it is possible to predict the stability of the process through the viscosity ratio at the interface and the normal stress difference ratio.

In the present work, the non-isothermal coextrusion fiber spinning with flow-induced crystallization is analyzed. The present model and the numerical simulation is an extension of the formulation originally proposed by Ching-China and Jyh-Chau (1996) in which Newtonian and PTT fluids are considered the core and the skin layer, respectively. The crystallization is taken into account using the kinetic equation used by Sung *et al.* (2005). The analysis is limited to the draw down region, where the fluids are stretched to form a thin fiber; the inertia, gravitational and surface tension are neglected. The influence of the polymeric flow rate fraction, the PTTs parameters model, the Deborah number and the flow induced crystallization is analyzed.

2. MATHEMATICAL MODEL

2.1 Assumptions

A simplified scheme of the co-extrusion fiber process is shown in Fig. 1. According to material properties and process conditions such as; incompressible and viscous fluid, low Reynolds number (ratio of inertial forces to viscous forces) and low weight material. The main assumptions of the model are: i) steady-state and nonisothermal flow, ii) the core and skin layer are Newtonian and PTT fluids, respectively, iii) crystallization kinetics for the skin layer is considered, iv) viscous and viscoelastic force are included and v) gravity, inertia and sur-face tension are neglected.



Fig. 1. Schematic diagram of the co-extrusion spinning process.

2.2 Governing Equations

According to the above assumptions, the mass and momentum equations are:

$$\frac{\partial \left(AV_z\right)}{\partial z} = 0,\tag{1}$$

$$\frac{\partial}{\partial z} \left[\phi A \left(\sigma_{zz}^{s} - \sigma_{rr}^{s} \right) + (1 - \phi) \left(\sigma_{zz}^{c} - \sigma_{rr}^{c} \right) \right] = 0.$$
 (2)

The first term in Eq. (2) is the axial tension contributed by PTT skin fluid while the second term is caused by the Newtonian core layer.

2.3 Constitutive Equations

For the Newtonian core layer, the stress tensor σ^c is proportional to the strain rate **D** as follows:

$$\sigma^c = -2\mu D \tag{3}$$

The radial and axial stress tensor components are, respectively

$$\sigma_{rr}^{c} = -\mu^{c} \frac{\partial V_{r}}{\partial r}, \qquad \sigma_{zz}^{c} = -2\mu^{c} \frac{\partial V_{z}}{\partial z}.$$
 (4)

The PTT constitutive equation, known for its robustness and accuracy in describing extensional deformation processes, is employed for the skin layer. The exponential PTT model is:

$$\exp(\frac{\varepsilon}{E}tr\sigma^{s})\sigma^{s} + \lambda\left((V.\nabla)\sigma^{s} - L.\sigma^{s} - \sigma^{s}.L\right) = 2\mu^{s}D,$$
(5)

$$tr\sigma^{s} = \sigma_{rr} + \sigma_{\theta\theta} + \sigma_{zz} = 2\sigma_{rr} + \sigma_{zz}, \qquad (6)$$

$$\mathbf{L} = \nabla \mathbf{V} - \boldsymbol{\xi} \mathbf{D},\tag{7}$$

where *E*, tro^s, λ and L are the elastic modulus, the trace of stress tensor, the relaxation time and the velocity gradient tensor, respectively. The dimension-less parameters ε and ξ represent the extensional and the shear thinning/thickening effects, respectively.

In the case of steady state uniaxial flow, Eq. (5) for radial and axial stresses is Ching-China and Jyh-Chau (1996):

$$\sigma_{rr} \exp(\frac{\varepsilon\lambda}{\mu^{s}}(\sigma_{zz} + 2\sigma_{rr})) + \lambda (V_{z} \frac{\partial\sigma_{rr}}{\partial z} + (1 - \xi)\frac{\partial V_{z}}{\partial z}\sigma_{rr}) = -\mu^{s}\frac{\partial V_{z}}{\partial z}.$$
(8)

$$\sigma_{zz} \exp(\frac{\varepsilon\lambda}{\mu^{s}}(\sigma_{zz} + 2\sigma_{rr})) + \lambda (V_{z} \frac{\partial\sigma_{zz}}{\partial z}) - 2(1-\xi)\frac{\partial V_{z}}{\partial z}\sigma_{zz} = 2\mu^{s}\frac{\partial V_{z}}{\partial z}.$$
(9)

2.4 Energy Equation

Considering absence of resistance to radial heat transfer by conduction between layers and in the axial direction but accounting the heat exchange be-tween skin layer and surrounding and the crystallization kinetic the energy equation is:

$$V_{z}\frac{\partial T}{\partial z} = -\frac{2\sqrt{\pi}h}{\rho C_{p}\sqrt{A}} \left(T - T_{a}\right) + \frac{\Delta H_{f}}{C_{p}} \left(V_{z}\frac{\partial X}{\partial z}\right), (10)$$

where ΔH_f , h, T_a , X, C_p and ρ are the heat of crystallization per unit mass, the heat transfer coefficient, the environment temperature, the crystallinity, the heat capacity and the density of skin fluid, respectively.

2.5 Crystallization Equation

For the crystallization kinetics, the model used by Joo *et al.* (2002) and Sung *et al.* (2005) is implemented here. In this model the crystallization rate is a function of temperature and of the molecular orientation. Then, in uniaxial extensional flow this molecular

orientation is related to the first normal stress difference as.

$$V_{z} \frac{\partial X}{\partial z} = n \left(\ln \left(\frac{X_{\infty}}{X_{\infty} - X} \right) \right)^{\frac{n-1}{n}} (X_{\infty} - X) K_{\max}$$

$$\exp(-4 \ln 2 \left(\frac{T - T_{\max}}{D} \right)^{2} + \kappa \left(\frac{\sigma_{zz} - \sigma_{rr}}{E} \right)),$$
(11)

where X and X_{∞} are the crystallinity and the maximum crystallinity, respectively. T_{max} is the fluid temperature at the maximum crystallization rate, K_{max} is the maximum crystallization rate, κ is the flow-induced crystallization factor and n is the Avrami index that in the case of uniaxial extension is one.

The set of Eqs. (1, 2, 8, 9, 10 and 11) is placed in dimensionless form using the following scaling parameters *a*, *v*, τ_{ii} , *x*, θ , θ_a , v_y , k_m , *d* and ζ defined as:

$$\begin{aligned} a &= \frac{A}{A_0}, \quad v = \frac{V_z}{V_0}, \quad \tau_{ii} = \frac{\sigma_{ii}A_0}{F}, \quad \chi = \frac{X}{X_\infty}, \quad \theta = \frac{T}{T_0}, \\ \theta_a &= \frac{T_a}{T_0}, \theta_{\max} = \frac{T_{\max}}{T_0}, k_m = \frac{k_{\max}L}{V_0}, d = \frac{D}{T_0}, \zeta = \frac{z}{L}. \end{aligned}$$

2.6 Dimensionless Equations

$$\frac{dav}{d\zeta} = 0, \tag{12}$$

$$\frac{d}{d\zeta} \left[a\phi N + 3(1-\phi)aGM \frac{dv}{d\zeta} \right] = 0$$
(13)

$$\tau_{zz} \exp\left\{\frac{\varepsilon De}{G}(3\tau_{zz} - 2N)\right\} +$$

$$De\left\{\frac{\sqrt{d\tau_{zz}}}{\sqrt{d\tau_{zz}}} - 2(1 - \varepsilon)\tau_{zz}\frac{dv}{dv}\right\} = 2G\frac{dv}{dv}.$$
(14)

$$\sum_{i=1}^{N} \left\{ \frac{\partial De}{\partial \zeta} - 2(1-\zeta) l_{zz} \frac{\partial \zeta}{\partial \zeta} \right\} + \sum_{i=1}^{N} \left\{ \frac{\partial De}{\partial \zeta} (3\tau_{zz} - 2N) \right\} + \sum_{i=1}^{N} \left\{ \frac{\partial De}{\partial \zeta} \right\}$$

$$De\left\{v\frac{dN}{d\zeta} + (1-\xi)N\frac{dv}{d\zeta} - 3(1-\xi)\tau_{zz}\frac{dv}{d\zeta}\right\} = 3G\frac{dv}{d\zeta}$$
(15)

$$v\frac{d\theta}{d\zeta} = -S_t a^{\frac{-1}{2}}(\theta - \theta_a) + \Delta H_f^*(v\frac{dx}{d\zeta}), \qquad (16)$$

$$v \frac{dx}{d\zeta} = (1-x)k_m$$

$$\exp\left[-4\ln 2\left(\frac{\theta - \theta_{\max}}{d}\right)^2 + \frac{\kappa NDe}{G}\right],$$
(17)

Where

$$\Delta H_f^* = \Delta H_f X_\infty \,/\, C_p T_o, \ N = \tau^s_{zz} - \tau^s_{rr}, \ M =$$

 $\mu^c / 2\mu_o^s$ and $G = \mu^s A_0 V_0 / FL$ are; the dimension-less crystallization heat, the first normal stress difference, the ratio of the core and skin viscosity in the fiber and the dimensionless axial force, respectively. Finally $S_t = 3.55hL / \rho C_p V_0 A_0^{1/2}$ is Stanton number and $De = \lambda V_0 / L$ is Debora number.



Fig. 2. Velocity profile as a function of dimension-less position for (a) extensional and (b) extensional+ shear deformations.

2.7 Boundary Conditions

The dimensionless bounday conditions for velocity, cross area, first normal stress difference, axial stress, temperature and crystallinity at the beginning and at the end of the fiber are:

$$\begin{array}{l} at \ \ \zeta = 0; \\ a = 1, v = 1, N = 1, \tau_{zz} = t_0, \theta = 1, x_c = 0 \quad (18) \\ at \ \ \zeta = 1; \ \ v = D_r = \frac{V_L}{V_0}, \end{array}$$

where D_r is the draw ratio indicating the relation between

the take up velocity V_L and the velocity at the beginning of the fiber V_0 . D_r determines the final condition for all the others variables, with this procedure reduce the order and grade of the differential equation for the velocity and the Eqs. (12-13) are integrated:

$$av = 1, \tag{19}$$

$$N = \frac{v}{\phi} - \frac{3(1-\phi)}{\phi} GM \frac{dv}{d\zeta},$$
(20)

The value of integration constants Eqs. (19-20) are set to unity (there is no loss of generality in the solution) Ching-China and Jyh-Chau (1996). Substitution of Eqs. (19-20) into Eqs. (14) and (17) is made. A set of simultaneous first-order ordinary differential equations is obtained; this set is solved by numerical procedures Shoichiro (2001):



Fig. 3. Extensional rate profile as a function of dimensionless position for (a) extensional and (b) extensional + shear deformations

$$\frac{dv}{d\zeta} = w,$$
(21)

$$\frac{dw}{d\zeta} = \frac{\exp(B)}{3(1-\phi)MGDe} - \frac{w}{v} \left[\frac{\exp(B)}{De} + (1-\xi)w \right] - \frac{w}{(1-\phi)M} \left[\frac{\phi}{2vDe} - \left(\frac{2-\xi}{3G}\right) + \left(\frac{\phi(1-\xi)\tau_{zz}}{vG}\right) \right]$$
(22)

$$\frac{d\tau_{zz}}{d\zeta} = \left[2G + 2De(1-\zeta)\tau_{zz}\right]\frac{w}{vDe} - \frac{\tau_{zz}\exp(B)}{vDe}, (23)$$

$$\frac{d\theta}{d\zeta} = -S_t v^{\frac{-1}{2}} (\theta - \theta_a) + \Delta H_f^* \left(\frac{d\chi}{d\zeta}\right), \tag{24}$$

(25)

$$= \frac{(1-\chi)}{v} k_m \exp\left[-4\ln 2\left(\frac{\theta - \theta_{\max}}{d}\right)^2 + \frac{\left(\frac{\nu}{\phi} - \frac{3(1-\phi)wMG}{\phi}\right)\kappa De}{G}\right]$$

Where $B = \frac{De}{G}\left[3\tau_{zz} - \frac{2\nu}{\phi} + \frac{6(1-\phi)6Mw}{\phi}\right]$ and
the corre-sponding boundary condition for Eq
 $w = \frac{1}{3MG}$ at $\zeta = 0$ Ching-China and Jyh-Chau

(1996).

 $\frac{d \chi}{d \zeta}$

3. **RESULTS AND DISCUSSIONS**

In this section, the numerical predictions of the non-isothermal co-extrusion fiber spinning with flow-induced crystallization are presented. The influence of the PTT's parameters model ($\varepsilon, \xi = 0$), the polymeric flow rate fraction and the flow induced crystallization on the main variables are analyzed.

3.1 Solution Method

The set of Eqs. (21)-(25) along with the corresponding boundary conditions (Eq. 18) are solved numerically using the shooting method Shoichiro (2001).

3.2 Velocity Profile

Figure 2 shows the effect of the polymeric flow rate fraction on the velocity profile. In Fig. 2a, when ($\varepsilon = 0, \xi = 0$) the Upper Convected Maxwell (UCM) is recovered, notice that increasing ϕ makes the velocity profile becomes linear, this behavior is associated with the material elasticity. The upper and lower limits ($\phi = 1$ and $\phi = 0$) correspond to elastic and viscous contributions of the material, respectively Denn (2008), Bird *et al.* (1987).

Similar numerical results were reported by Ching-China and Jyh-Chau (1996) using the UCM for the mixture PET/PP. The case of extensional deformation ($\varepsilon = 0.015, \xi = 0$) and varying ϕ , the same behavior is obtained but in lower magnitude except for a viscous fluid $\phi = 0$. The Fig. 2b show the cases of shear ($\varepsilon = 0, \xi = 0.5$) and extensional/shear deformations ($\varepsilon = 0.015, \xi = 0.5$), all the velocity curves are closer and similar of the results reported by Ching-China and Jyh-Chau (1996) for the polymeric isothermal co-extrusion, the shear thinning effect is dominant in the PTT model and instability phenomena could happen for higher extensional rates.

3.3 Extensional Rate Profile

In Fig. 3 shows the extensional rate as a function of the axial position for different values of polymeric flow rate fraction, Fig. 3a shows that lower extensional rate is the UCM with $\phi = 1$ and higher extensions for ($\varepsilon =$ $0.015, \xi = 0$) reducing ϕ . At the beginning of the fiber there is a reduction of the ex-tensional rate, which can be explained as a yield stress or the required energy for the material starts to flow, after this point a linear behavior for an elas-tic material with small extension and higher deformations at the end of the fiber for an extensional viscous fluid are exhibit. Considering both deformations extensional/shear ($\varepsilon = 0.015, \xi = 0.5$) the result is a single curve with separation at the end of the fiber Fig. 3b.

The effect of the elastic force reduces the velocity because the material tends to recover its original form and part of the total energy of the system is used in this process, the viscous case all the energy is employed in diffusional process.

3.4 Axial Stress Tensor

In Fig. 4 the axial stress tensor T zz as a function of the position is presented, for the UCM with $\phi = 1$, T zz increases up to reach a constant value at the beginning of the process, it is known that UCM reports an unreal behavior for large deformations. The polymeric flow rate fraction has strong influence in this process Fig. 4a, Takeshi et al. (1996) used the UCM with $\phi = 0.5$ for the ratio 5/5 PET/PP system, They reported the same behavior of this work. A significant increase of T zz can be obtained incorporating the extensional parameter in the PTT model, if shear deformation is also included, the later dominates the process and modifies the behavior to a single curve reaching higher tensile stresses, see Fig. 4b.



Fig. 4. Axial stress as a function of dimensionless position for (a) extensional and (b) extensional + shear deformations

High-speed non-isothermal melt spinning is associated with a concentrated neck-like deformation process and the development of high tensile stresses that result in the so-called flow (or axial stress) induced crystallization (FIC). In Fig. 5 shows the crystallization percentage as a function of the axial position varying the polymeric flow rate fraction ϕ and the crystallinity parameter control κ . For $\kappa = 0$, no induced crystallization is considered, however the crystallinity increases up to reach a constant value Fig. 5a. Doufas et al. (2000) published similar results for a Nylon-66 fiber, They reported that the crystallinity is dependent on the polymeric flow rate. Extensional deformation augmnted the crystallinity percentage keeping the behavior Figs. 5a-5c. same



Fig. 5. Crystallization profile as a function of dimensionless position for extensional deformation

Increasing κ the crystallization pro-cess is faster but its percentage is reduced, except for a viscous fluid where the crystallinity percentage is augmeted Figs. 5a-5c. In Fig. 6 the crystallinity profile for a extensional/shear deformation is shown, the crystallinity porcentage increases but the shear thinning effect dominates the process due to all the curves are closer varying the polymeric flow rate fraction κ except for a viscous fluid where is sensible to this process.

3.6 Temperature Profile

The temperature profile as a function of the axial position without crystallization is shown in Fig. 7. The temperature decreases monotonically along the fiber due to the heat transfer by convection mechanism, this effect is considered in the model by the Stanton dimensionless number.



Fig. 6. Crystallization profile as a function of dimensionless position for extensional/shear deformation3.5 Crystallization Profile

In Figs. 7a and 7b all curves have the same behavior, the temperature along the fiber is dependent on the type of deformation and on the polymeric flow rate fraction. Takeshi *et al.* (1996) using a non-isothermal model reported similar results. Including the shear thinning effect, the temperature decrease monotonically having the same behavior of the previous one, but the curves are closer indicating the strong influence of the shear parameter $\xi = 0.5$ and more viscous dissipation is generated Fig. 7b.

The temperature profile taking into account the crystallinity kinetic process is shown in Fig. 8. In this figure exhibits anincrement of the temperature due to crystallization effect, this behavior is reported in the numerical predictions of polymeric fiber extrusion by Eder *et al.* (1990), McHugh (1990). A strong influence of

the stress induce crys-tallization on the temperature profile is at the be-ginning of the process, see Figs 8a and 8b. The higher increment of the temperature is reached when the process takes into account a shear thinning fluid and the stress induced crystallization Fig. 9. After this process the temperature decreases monotonically due to the convection heat transfer to the environment



Fig. 7. Temperature profile as a function of dimensionless position for (a) extensional and (b) extensional + shear deformations

4. CONCLUSIONS

The numerical simulation of the non-isothermal steady co-extrusion fiber spinning with flowinduced crystallization is performed. The formulation originally proposed by China *et al.* is extended, Newtonian and PTT fluids are considered the core and the skin layer, respectively.

The numerical results of the non-isothermal fiber spinning process without crystallization but taking into account the extensional and shear thinning/thickening deformation, exhibit the same behavior of the isothermal case reported by Ching-China and Jyh-Chau (1996). The extension rate, tensile stress tensor and temperature profiles are in-fluenced by the polymeric flow rate fraction and thinning parameters of the PTT model.

Higher axial stress tensor are reached when the shear thinning parameter is included in the PTT model, the shear thinning effect dominates the process, obtaining a single curve. The extensional rate as a function of the position shows a linear behavior for high elastic fluid, this behavior is lost when viscous contribution is increased.

The extensional rate, stress tensor and temperature profiles are affected when the flowinduced crystallinity. The degree of crystallinity profile show a monotonic increased up to reach a constant value. On the other hand, the tensile stress has a strong influence on the degree of crystallinity at the be-ginning of the fiber extruded, the zone where the crystallinity is constant depend on the viscoelastic flow fraction.







Fig. 9. Crystallization profile as a function of dimensionless position for extensional + shear deformations varing the crystallinity parameter κ

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