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Effect of die exit stress state, Deborah number, uniaxial and planar extensional rheology on the neck-in phenomenon in polymeric flat film production

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A B S T R A C T

In this work, effect of the second to first normal stress difference ratio at the die exit, $-N_2/N_1$, uniaxial extensional strain hardening, $\frac{\eta_{E,U,max}}{3\eta_0}$, planar-to-uniaxial extensional viscosity ratio, $\frac{\eta_{E,P}}{\eta_{E,U}}$, and Deborah number, *De*, has been investigated via viscoelastic isothermal modeling utilizing 1D membrane model and a single-mode modified Leonov model as the constitutive equation. Based on the performed parametric study, it was found that there exists a threshold value for *De* and $\frac{\eta_{E,U,max}}{3\eta_0}$, above which, the neck-in starts to be strongly dependent on $-N_2/N_1$. It was found that such critical *De* decreases if $-N_2/N_1$, $\frac{\eta_{E,U,max}}{3\eta_0}$ increases and/or $\frac{\eta_{E,P}}{\eta_{E,U}}$ decreases. Numerical solutions of the utilized model were successfully approximated by a dimensionless analytical equation relating the normalized maximum attainable neck-in with $\frac{\eta_{E,U,max}}{3\eta_0}$, $\frac{\eta_{E,P}}{\eta_{E,U}}$, $-N_2/N_1$ and *De*. Suggested equation was tested by using literature experimental data considering that $-N_2/N_1$ depends on die exit shear rate, temperature and utilized constitutive model parameters for given polymer melt. It was found that approximate model predictions are in a very good agreement with the corresponding experimental data for low as well as very high Deborah numbers, at which neck-in strongly depends on $-N_2/N_1$. It is believed that the obtained knowledge together with the suggested simple model can be used for optimization of the extrusion die design (influencing flow history and thus die exit stress state), molecular architecture of polymer melts and processing conditions to suppress neck-in phenomenon in production of very thin polymeric flat films.

1. Introduction

The extrusion film casting technology is an industrially important process that has a firm position on the market due to its capability to produce high quality thin polymeric films at high production rates. Those films can be used in different applications such as wrapping materials, barriers reducing permeability for air and vapor, or as a separator membrane for rechargeable batteries in mobile devices and electric vehicles.

In this technology, the polymer melt is extruded through a slit die to form a thick sheet that is subsequently intensively stretched in the axial direction, hauled off and quenched by a rotating drum stabilizing the film dimensions. [1,2] (Fig. 1). Except of an initial swelling, the thickness of the film decreases monotonically due to high Draw ratio (the haul off speed divided by the die exit velocity). In such a case, film width is gradually reduced toward the stretching/cooling roll, which is called neck-in phenomenon.

Aside from that, the interrelated defect of edge-beads promotes the lateral portion of the film to being substantially thicker than its central part (Fig. 2). While the first phenomenon affects a production rate, the second calls for a post-production trimming since solely central portion of the film is uniform in thickness. Understanding the relationship between material parameters and processing conditions including the flow in the die might be the way to effectively control the extent of neck-in and edge-beading as even a small reduction of these defects may bring increased efficiency considering high production rates in this manufacturing process.

Scientific investigation of the extrusion film casting process has been addressed in many works dealing with both a steady and transient approaches to the problem. Initial studies were dedicated to an investigation of the hydrodynamic instability observed during the production of fibers called draw resonance [3–5] and then expanded for films in [6] where the numerical modeling for film casting using the one-dimensional isothermal model of Newtonian fluid was utilized for

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List of symbols

A	aspect ratio (1)
A_1, A_2	fitting parameters of analytical model (1)
b	dissipation term (s^{-1})
$\bar{b}$	dimensionless dissipation term (1)
$\underline{c}$	recoverable Finger tensor (1)
$\underline{c}^{-1}$	inverse recoverable Finger tensor (1)
$\overset{0}{\underline{c}}$	jaumann (corotational) time derivative of the recoverable Finger strain tensor (s^{-1})
c_{xx}	normal component of the recoverable Finger tensor in axial x-direction (1)
c_{yy}	normal component of the recoverable Finger tensor in transverse y-direction (1)
c_{zz}	normal component of the recoverable Finger tensor in thickness z-direction (1)
$\underline{D}$	deformation rate tensor (s^{-1})
De	Deborah number (1)
DR	draw ratio (1)
$\underline{e}_p$	irreversible rate of strain tensor (s^{-1})
E	dimensionless take-up force (1)
E_a	flow activation energy ($J{\cdot}mol^{-1}$)
e	half-thickness of the film at any x location (mm)
e_0	die half-gap (half-thickness of the film at the die exit) (mm)
$\bar{e}$	dimensionless half-thickness of the film at any x location (1)
F	take-up force (stretching force) (N)
f	rate of deformation in transverse y-direction (s^{-1})
G	linear Hookean elastic modulus (Pa)
g	rate of deformation in thickness z-direction (s^{-1})
i	index i, noting the spatial direction (1)
$I_{1,\,c}$	first invariant of recoverable Finger tensor (1)
$I_{2,\,c}$	second invariant of recoverable Finger tensor (1)
j	index j (1)
L	half-width of the film at any x location (mm)
L_0	half-width of the die (half-width of the film at the die exit) (mm)
$\bar{L}$	dimensionless half-width of the film at any x location (1)
MFI	melt flow index (g/10 min)
MFR	mass flow rate ($kg{\cdot}h^{-1}$)
M_n	number average molar mass ($g{\cdot}mol^{-1}$)
M_w	mass average molar mass ($g{\cdot}mol^{-1}$)
NI	maximum attainable neck-in (mm)
NI^*	normalized maximum attainable neck-in (1)
N_1	first normal stress difference (Pa)
N_2	second normal stress difference (Pa)
n	non-linear Leonov model parameter (1)
n_0	non-Newtonian index (1)
n_s	number of sample points (1)
Q	volumetric flow rate ($m^3{\cdot}s^{-1}$)
R	gas constant ($J{\cdot}K^{-1}{\cdot}mol^{-1}$)
T	melt temperature at the die (°C)
T_0	reference temperature in the Arrhenius law (°C)
u	axial velocity component of the film at any x location ($mm{\cdot}s^{-1}$)
$u(X)$	chill roll speed ($mm{\cdot}s^{-1}$)
u_0	axial velocity component at the die exit ($mm{\cdot}s^{-1}$)
$\bar{u}$	dimensionless axial velocity component of the film at any x location (1)
v	transverse velocity component of the film at any x location ($mm{\cdot}s^{-1}$)
W	elastic potential (Pa)
w	thickness velocity component of the film at any x location ($mm{\cdot}s^{-1}$)
X	take-up length (stretching distance, air gap) (mm)
x	position in axial x-direction (mm)
$\bar{x}$	dimensionless position in axial x-direction (1)
y_i	observed value (1)
$\hat{y}_i$	predicted value (1)
x, y, z	spatial coordinates in axial, transverse and thickness direction, respectively (1)
Z_x, Z_y, Z_z	substitution variables (1)
$\left(\frac{du}{dx}\right)_M$	mean value of extensional strain rate in the air gap (s^{-1})
$\frac{dc_{xx}}{d\bar{x}}$, $\frac{dc_{yy}}{d\bar{x}}$, $\frac{dc_{zz}}{d\bar{x}}$	derivative of Finger tensor components with respect to dimensionless $\bar{x}$ position (1)
$\frac{d\bar{u}}{d\bar{x}}$, $\frac{d\bar{L}}{d\bar{x}}$, $\frac{d\bar{e}}{d\bar{x}}$	derivative of dimensionless axial, transverse and thickness velocity component with respect to dimensionless $\bar{x}$ position (1)

Greek symbols

α	Arrhenius law parameter (K)
α_1, α_2	fitting parameters of analytical model (1)
β	non-linear Leonov model parameter (1)
$\dot{\gamma}_{COR}$	corrected shear rate by Rabinowitsch correction for the rectangle channel (s^{-1})
$\underline{\delta}$	unit tensor (Kronecker delta) (1)
δ	δ shift function (1)
η_0	Newtonian viscosity (Pa·s)
$\eta_{E,\,P}$	steady planar extensional viscosity (Pa·s)
$\eta_{E,P,\max}$	maximal steady planar extensional viscosity (Pa·s)
$\eta_{E,\,U}$	steady uniaxial extensional viscosity (Pa·s)
$\eta_{E,\,U,\max}$	maximal steady uniaxial extensional viscosity (Pa·s)
θ	fitting parameters of analytical model (1)
λ	relaxation time (s)
ν	non-linear Leonov model parameter (1)
ξ	non-linear Leonov model parameter (1)
ρ	polymer density ($g{\cdot}cm^{-3}$)
$\underline{\tau}$	extra stress tensor (Pa)
τ_{xx}	normal stress in axial x-direction (Pa)
τ_{yy}	normal stress in transverse y-direction (Pa)
τ_{zz}	normal stress in thickness z-direction (Pa)
$\bar{\tau}_{xx}$	dimensionless normal stress in axial x-direction (1)
$\bar{\tau}_{yy}$	dimensionless normal stress in transverse y-direction (1)
$\bar{\tau}_{zz}$	dimensionless normal stress in thickness z-direction (1)
φ_1, φ_2	fitting parameters of analytical model (1)
ψ_1, ψ_2, ψ_3, ψ_4	fitting parameters of analytical model (1)

the first time. Other authors followed and employed different constitutive equations for power-law [7], and viscoelastic fluids using modified convected-Maxwell [8] and modified Giesekus model [9,10]. Due to the assumed kinematics for the free surface flow at the drawing zone, the model could not capture edge-bead defect and contraction in film width.

First efforts to overcome this limitation and to accommodate ability to predict neck-in were made for a Newtonian fluid at isothermal [11] and non-isothermal conditions [12–14]. Lately, improved isothermal two-dimensional membrane model having the capability to capture the development of edge-beads under the stationary conditions was released; isothermal Newtonian [15] and viscoelastic Maxwell and Giesekus fluid [16], and models that take thermal effects into account for Newtonian [17] and viscoelastic Larson fluid [18]. In the meantime, simplified one-dimensional membrane approach based on a supplementary kinematic hypothesis, that originally brought for a float glass