Variational Principle based Modeling of Film Blowing Process for LDPE Considering Non-Isothermal Conditions and Non-Newtonian Polymer Melt Behavior

ROMAN KOLARIK\textsuperscript{a,b} and MARTIN ZATLOUKAL\textsuperscript{a,b}
\textsuperscript{a}Centre of Polymer Systems, University Institute
Tomas Bata University in Zlin
Nad Ovcirnou 3685, 760 01 Zlin
CZECH REPUBLIC
\textsuperscript{b}Polymer Centre, Faculty of Technology
Tomas Bata University in Zlin
TGM 275, 762 72 Zlin
CZECH REPUBLIC
rkolarik@ft.utb.cz \textsuperscript{a}http://web.uni.utb.cz/, \textsuperscript{b}http://web.ft.utb.cz/

Abstract: In this work, film blowing process analysis has been performed theoretically by using minimum energy approach for non-Newtonian polymer melts considering non-isothermal processing conditions and the obtained predictions were compared with both, theoretical and experimental data (bubble shape, velocity and temperature profiles) taken from the open literature. It has been found that model predictions are in very good agreement with the corresponding experimental data.

Key-Words: Mathematical modeling, Variational principle, Numerical analysis, Non-isothermal film blowing, Non-Newtonian fluids, Films.

1 Introduction
The film blowing process belongs to one of the most commonly used industrial processes producing biaxially oriented films applicable to daily used products in the food processing industry, waste industry and in the medical industry as well [1].

In the film blowing process a polymer melt is extruded at a constant flow rate through an annular die to a continuous tube which is consequently stretched and inflated by the take-up force and internal bubble pressure. At the same time, the raising bubble is cooled by an air ring with/without an internal bubble cooling system IBC. Then, above the freezeline height, the solid bubble is stabilized by a calibration cage and consequently is transformed to a lay-flat film shape by the collapsing frames. Finally, with the help of the nip rolls, the film is drawn to a wind up roll (see Fig. 1).

In order to understand the relationship between the die design, rheology and processing parameters, modeling of the film blowing process is necessary. The first film blowing model was developed in 1970 by Pearson and Petrie [2, 3] who assumed isothermal process conditions for Newtonian polymer behavior. In their work, the film is viewed as a thin shell in tension in the machine and circumferential direction. This model became the basis of the most following film blowing models which are summarized in the work of Muke et al. [4]. In 2004, Zatloukal and Vlcek [5] brought a new view on film blowing modeling by using variational principle to obtain the bubble shape which satisfies minimum energy requirements. This model is taking into account non-isothermal processing conditions and non-Newtonian polymer behavior [6]. The latest
models are introduced by the Muslet and Kamal [7] who applied the Phan-Thien and Tanner (PTT) constitutive equation in the liquid region and neo-Hookean model in the solid region with assuming that crystallization effects and variable heat transfer coefficient are counted. Beaulne and Mitsoulis [8] used K-BKZ integral constitutive equation with a spectrum of relaxation times. Sarafrazi and Sharif [9] applied extended Pom-Pom (XPP) model with non-isothermal flow and with the Nakamura equation for crystallization kinetics.

The aim of this paper is to verify predicting capabilities of the variational principle based Zatloukal/Vlcek film blowing model, taking non-isothermal processing conditions and non-Newtonian polymer behavior into account, on Tas’s PhD thesis experimental data [10] and consequently compare the model results with the Muslet/Kamal model predictions [7].

2 Mathematical Modeling

With the Zatloukal/Vlcek variational principle based formulation [5], the bubble of the film blowing process is introduced as an elastic membrane which is deformed due to the load, \( p \), and the take-up force, \( F \), in such a way that the resulting stable bubble shape satisfies minimum energy requirements. This Zatloukal/Vlcek formulation [5], extended for the non-isothermal processing conditions and non-Newtonian polymer behavior [6, 11], is given by the complete set of equations, summarized in Tables 1 and 2, and applied for a development of the numerical scheme (see Fig. 2) described in detail in [6].

As can be seen in Fig. 3, the final results of the numerical scheme (bubble shape, velocity and temperature profiles, take-up force, \( F \), and internal bubble pressure, \( \Delta p \)) are reached when the previous and the newly calculated value of the velocity at the freezezine height, viscosity and the internal bubble pressure are constant.

It also should be mentioned that two numerical schemes are developed and more described in [6]. In the first one, bubble shape, \( pJ \), was fixed and \( \Delta p, F \) were unknown parameters whereas in the second one internal bubble pressure, \( pJ \), was fixed and bubble shape, \( pJ \), and take-up force, \( F \), were taken as unknown variables (see Fig. 3).

Table 1. A full set of the Zatloukal/Vlcek model equations employing non-isothermal processing conditions and non-Newtonian polymer behavior (the meaning of the used symbols is described at the end of this paper in “List of symbols”).

<table>
<thead>
<tr>
<th>The Zatloukal/Vlcek Formulation</th>
</tr>
</thead>
<tbody>
<tr>
<td>( E_n = \frac{p}{2} \left( y' dx - p \right) J dx )</td>
</tr>
<tr>
<td>( J = \frac{1}{2} F(y' - py') + \lambda \left( y' \right)^2 )</td>
</tr>
<tr>
<td>( \frac{\partial J}{\partial y'} - \frac{\partial J}{\partial y} \frac{\partial y}{\partial y'} = 0 )</td>
</tr>
<tr>
<td>( F_J - \frac{1}{2} J + p = 0 )</td>
</tr>
<tr>
<td>( J = \frac{1}{2} \pi \rho_1 )</td>
</tr>
<tr>
<td>( y(x = L) = R_2, \text{BUR} = R_2 )</td>
</tr>
<tr>
<td>( y = \left( R_1 - \rho \right) \cos \left( \frac{\pi}{L} x \right) - \rho_y \left( \rho \right) \sin \left( \frac{\pi}{L} x \right) )</td>
</tr>
<tr>
<td>( F = \frac{1}{2p \rho} )</td>
</tr>
<tr>
<td>( \alpha' = \left[ \frac{2p \rho - R_2 \text{BUR} R_2}{p \rho - \text{BUR} R_2} \right] )</td>
</tr>
<tr>
<td>( \Delta p = \frac{p \rho}{2\pi} \frac{1}{\pi \rho} )</td>
</tr>
<tr>
<td>( F_{\text{wall}} = \left</td>
</tr>
</tbody>
</table>

The Continuity Equation

\( Q = 2 \rho n \left( x, h(x), y(x) \right) \) \( \rho = 0.934 \cdot 0.901 \cdot 10^{15} \cdot 10^{15} \cdot 0.875 \)

The Constitutive Equation

\( \tau = 2 \left[ \frac{1}{3} \dot{\gamma} \left( \dot{e} \right) \text{H}_{\dot{e}} \frac{\dot{e}}{\dot{e}} \right] \)

\( \sigma \left( \dot{e} \right) \left( \dot{e} \right) \text{H}_{\dot{e}} \frac{\dot{e}}{\dot{e}} = \lambda \left[ \frac{1}{3} \dot{\gamma} \left( \dot{e} \right) \text{H}_{\dot{e}} \frac{\dot{e}}{\dot{e}} \right] \)

\( \left( \dot{e} \right) \left( \dot{e} \right) \text{H}_{\dot{e}} \frac{\dot{e}}{\dot{e}} = \gamma \left( \dot{e} \right) \frac{\dot{e}}{\dot{e}} \frac{1}{\gamma \left( \dot{e} \right)} \frac{1}{\gamma \left( \dot{e} \right)} \)

\( \alpha_0 = \exp \left( \frac{E_1}{R \left( 273.15 + T \right)} \right) \)

The Internal Force at the Freezeline Height

\( F_n = \frac{2p \rho}{2 \pi \rho} \)

\( J = \frac{\Delta p}{2 \pi \rho} \)

The Energy Equation

\( \frac{\partial \epsilon}{\partial t} \frac{\dot{e}}{\dot{e}} = -2 \frac{p \rho}{m} \left[ H(\tau - T_{\alpha}) + \sigma \left( \frac{1}{\gamma \left( \dot{e} \right)} \right) \right] \frac{\dot{e}}{\dot{e}} \frac{1}{\gamma \left( \dot{e} \right)} \frac{1}{\gamma \left( \dot{e} \right)} \)
3 Results and Discussion

The predicting capabilities of the Zatloukal/Vlcek non-isothermal film blowing model, employing non-Newtonian polymer melts, were tested for the LDPE film blowing experiments provided in the Tas's Ph.D. thesis [10].

Firstly, the rheological characteristics of the LDPE material (L1 – experiment No. 18) taken from [10] were fitted by the generalized Newtonian model.

As can be seen in Fig. 2, the used generalized Newtonian model is able to describe extensional as well as shear viscosity data very well. Then, all model, material (LDPE L1 – experiment No. 18...
Tables 3-5 summarize the processing parameters for chosen processing conditions.

**Table 3. The Zatloukal/Vlcek film blowing model parameters.**

<table>
<thead>
<tr>
<th>BUR (-)</th>
<th>L (m)</th>
<th>Δp (Pa)</th>
<th>R₀ (m)</th>
<th>H₀ (m)</th>
<th>TUR (-)</th>
<th>m (kg s⁻¹)</th>
</tr>
</thead>
<tbody>
<tr>
<td>2.514</td>
<td>0.2057</td>
<td>95</td>
<td>0.0192</td>
<td>0.0008</td>
<td>4.803</td>
<td>0.001167</td>
</tr>
</tbody>
</table>

**Table 4. Generalized Newtonian constitutive equation parameters (A₁ = 1, α = 20).**

<table>
<thead>
<tr>
<th>η₀ (Pa s⁻¹)</th>
<th>λ (s⁻¹)</th>
<th>a (-)</th>
<th>n (-)</th>
<th>α (-)</th>
<th>β (-)</th>
<th>ζ (-)</th>
</tr>
</thead>
<tbody>
<tr>
<td>23,809</td>
<td>2.78</td>
<td>0.83</td>
<td>0.34</td>
<td>1.10⁻⁵</td>
<td>1.0910⁻⁷</td>
<td>0.04722</td>
</tr>
</tbody>
</table>

**Table 5. Temperature parameters.**

<table>
<thead>
<tr>
<th>Tᵣ (°C)</th>
<th>Tₑ (°C)</th>
<th>Tₛ (°C)</th>
<th>Tₑₑ (°C)</th>
<th>Eₐ (J mol⁻¹)</th>
<th>R (J K⁻¹ mol⁻¹)</th>
<th>Cᵥ (J kg⁻¹ K⁻¹)</th>
</tr>
</thead>
<tbody>
<tr>
<td>25</td>
<td>96</td>
<td>175</td>
<td>190</td>
<td>66,000</td>
<td>8.314</td>
<td>2,300</td>
</tr>
</tbody>
</table>

The following Figs. 4-6 were generated with the help of the stated parameters and the both numerical schemes. In more detail, the Zatloukal/Vlcek model predictions for the bubble shape (Fig. 4), velocity (Fig. 5) and temperature profiles (Fig. 6) are compared with Tas’s experimental data [10] together with theoretical predictions by Muslet/Kamal model [7]. Based on the Figs. 4-6 and Table 6, it is nicely visible that the predictions of the Zatloukal/Vlcek model are in very good agreement with the experimental data.

**Fig. 3. Iteration scheme of the Zatloukal/Vlcek film blowing model.**

**Fig. 4. Comparison of the bubble shapes between the proposed Zatloukal/Vlcek model prediction [5], experimental data for the LDPE L1 (experiment No. 18) taken from Tas’s Ph.D. thesis [10] and the Muslet/Kamal model prediction [7].**

**Fig. 5. Comparison of the velocity profiles between the proposed Zatloukal/Vlcek model prediction [5, 6], experimental data for the LDPE L1 (experiment No. 18) taken from Tas’s Ph.D. thesis [10] and the Muslet/Kamal model prediction [7].**
The overall results, i.e. Figs. 4-6 and Table 6, indicate that the predicted bubble shapes, velocity and temperature profiles as well as the predicted internal bubble pressures, take-up forces and stresses at the freezeline height by the Zatloukal/Vlcek model are in very good agreement with the Tas’s experimental data.

Table 6. Comparison between the Zatloukal-Vlcek [5, 6] model predictions (the calculated results for the fixed bubble shape $p_J$ and internal bubble pressure $\Delta p$ are provided in the parentheses and without parentheses, respectively) and Tas’s experimental data for LDPE material (L1-experiment No. 18) [10].

<table>
<thead>
<tr>
<th>Models</th>
<th>$\Delta p$ (Pa)</th>
<th>$F$ (N)</th>
<th>$\sigma_{11}$ (MPa)</th>
<th>$\sigma_{33}$ (MPa)</th>
</tr>
</thead>
<tbody>
<tr>
<td>Experimental data (Tas)</td>
<td>95</td>
<td>6.9</td>
<td>0.380</td>
<td>0.075</td>
</tr>
<tr>
<td>Zatloukal/Vlcek</td>
<td>95.000</td>
<td>4.922</td>
<td>0.245</td>
<td>0.069</td>
</tr>
<tr>
<td>(99.170)</td>
<td>(4.951)</td>
<td>(0.246)</td>
<td>(0.072)</td>
<td></td>
</tr>
<tr>
<td>Muslet/Kamal</td>
<td>data not available</td>
<td></td>
<td></td>
<td></td>
</tr>
</tbody>
</table>

4 Conclusion

In this paper, the predicting capabilities of the variational principle based Zatloukal/Vlcek film blowing model describing non-Newtonian polymer melts considering non-isothermal processing conditions have been successfully compared with the experimental data (bubble shape, velocity and temperature profiles, internal bubble pressure and take-up force) as well as with the theoretical predictions of the Muslet/Kamal model.

Acknowledgement

The authors wish to acknowledge the Grant Agency of the Czech Republic (grant No. P108/10/1325) and Operational Program Research and Development for Innovations co-funded by the European Regional Development Fund (ERDF) and national budget of Czech Republic, within the framework of project Centre of Polymer Systems (reg. number: CZ.1.05/2.1.00/03.0111) for the financial support.

List of symbols:

- $A$ Zatloukal-Vlcek model function (1)
- $A_1$ Generalized Newtonian model parameter (1)
- $a$ Generalized Newtonian model parameter (1)
- $a_T$ Arrhenius temperature shift factor (1)
- $a_{T,Bubble}$ Bubble temperature shift factor (1)
- $a_{TS}$ Mean value of the Arrhenius temperature shift factor $a_T$ (1)
- $a_T$ Average temperature shift factor (1)
- $BUR$ Blow-up ratio (1)
- $C_p$ Specific heat capacity (J kg$^{-1}$ K$^{-1}$)
- $D$ Deformation rate tensor (s$^{-1}$)
- $dx$ Element length in x direction (m)
- $E_a$ Activation energy (J/mol)
- $F$, $F_{total}$ Take-up force (N)
- $F_{local}$ Local film thickness (m)
- $H_T$ Heat transfer coefficient (W m$^{-2}$ K$^{-1}$)
- $H_0$ Bubble thickness at the die exit (m)
- $H_1$ Bubble thickness at the freezeline height (m)
- $h(x)$ Local film thickness (m)
- $\bar{h}$ Mean value of bubble thickness along the bubble (m)
- $\bar{I}_{[\rho]}$ First invariant of the absolute value of deformation rate tensor (s$^{-1}$)
- $\bar{I}_{[\rho]}$ Mean value of the first invariant of deformation rate tensor (s$^{-1}$)
- $II_D$ Second invariant of deformation rate tensor (s$^{-1}$)
- $\overline{II}_D$ Mean value of the second invariant of deformation rate tensor (s$^{-1}$)
- $III_D$ Third invariant of deformation rate tensor (s$^{-1}$)
- $\overline{III}_D$ Mean value of the third invariant of deformation rate tensor (s$^{-1}$)
- $I$ Potential energy functional (N)
J  Bubble compliance (Pa⁻¹)
L  Freezeline height (m)
$m$  Mass flow rate (kg s⁻¹)
$n$  Power-law index (1)
$\nu$  Internal load (Pa·m)
$Q$  Volumetric flow rate (m³·s⁻¹)
$R$  Universal gas constant (J·K⁻¹·mol⁻¹)
$R_0$  Die radius (m)
$R_2$  Bubble radius at the freezeline height (m)
$T$  Local bubble temperature (°C)
$T_{\text{air}}$  Air temperature (°C)
$T_{\text{die}}$  Die exit melt temperature (°C)
$T_{\text{ref}}$  Reference temperature (°C)
$T_s$  Average bubble temperature (°C)
$T_{\text{solid}}$  Solidification (freezeline) temperature (°C)
$T_{\text{air}}$  Air temperature (°C)
$T_{\text{solid}}$  Solidification (freezeline) temperature (°C)
$T_{\text{ref}}$  Reference temperature (°C)
$T_s$  Average bubble temperature (°C)
$\tau$  Extra stress tensor (Pa)
$\tau_{11}$  Extra stress in the machine direction (Pa)
$\tau_{22}$  Extra stress in the thickness direction (Pa)
$\phi$  Average absolute degree of crystallinity (1)
$\varphi$  Zatloukal-Vlcek model function (1)
$\psi$  Generalized Newtonian model parameter (1)
$\nabla v$  Velocity gradient tensor (s⁻¹)

Greek symbols:
$\alpha$  Generalized Newtonian model parameter (s)
$\beta$  Generalized Newtonian model parameter (1)
$\Delta H_f$  Heat of crystallization per unit mass (J/kg)
$\Delta p$  Internal bubble pressure (Pa)
$\dot{e}_1$  Extensional rate in machine direction (s⁻¹)
$\dot{e}_2$  Extensional rate in thickness direction (s⁻¹)
$\dot{e}_3$  Extensional rate in circumferential direction (s⁻¹)
$\varepsilon$  Emissivity (1)
$\overline{\dot{e}}_1$  Mean value of extensional rate in machine direction (s⁻¹)
$\overline{\dot{e}}_2$  Mean value of extensional rate in thickness direction (s⁻¹)
$\overline{\dot{e}}_3$  Mean value of extensional rate in circumferential direction (s⁻¹)
$\zeta$  Generalized Newtonian model parameter (1)
$\eta$  Viscosity (Pas)
$\eta_0$  Newtonian viscosity (Pas)
$\eta_1$  Average bubble viscosity (Pas)
$\lambda$  Relaxation time (s)
$\lambda_1$  Lagrange multiplier (Pa)
$\pi$  Ludolf’s number (1)
$\rho$  Polymer density (kg·m⁻³)
$\sigma_B$  Stefan-Boltzmann constant (W·m⁻²·K⁻⁴)
$\sigma_{11}$  Total stress tensor in machine direction (Pa)
$\sigma_{33}$  Total stress tensor in circumferential direction (Pa)

References: