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Stabilization of film casting by an encapsulation extrusion method

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Abstract

A film casting simulation has been used to demonstrate why the encapsulation extrusion coating process is so effective industrially in enhancing the stability of the extrusion coating of low-melt-strength polymers. In the present study, it is particularly intended to explain theoretically why and how the co-extrusion of high-melt-strength polymers like low-density polyethylene (LDPE)—attached at the periphery using encapsulation dies—dramatically improves the film coating process of low-melt-strength polymers like high-density polyethylene (HDPE). The undesirable neck-in and draw resonance phenomena frequently occurring in the extrusion coating of HDPE are shown in this study to be due to the fact that lower axial tension in the HDPE film rendered by its low melt strength leads to process instabilities. High melt strength LDPE will then increase the film tension when externally-attached, and as a result improves the stability of HDPE extrusion coating.

Keywords: Draw resonance; Encapsulation dies; Extrusion film coating; Film casting; Film thickness; Film width; High-density polyethylene (HDPE); Low-density polyethylene (LDPE); Neck-in; Stability

1. Introduction

The film casting process, widely used in the film industry, is a high-speed process for making oriented film. Polymer melts are extruded from a flat die, stretched and cooled by the pulling motion of the chill roll, resulting in film product with frozen-in molecular orientations [1–7]. The draw ratio is defined between the die and the chill roll, as the ratio of the take-up velocity of the film at the chill roll to its extrusion velocity at the die exit. When this draw ratio is increased beyond a certain critical value, the film casting process can become unstable, i.e. an instability called draw resonance occurs, in spite of constant extrusion and take-up speeds. In the unstable regions of the operating process conditions, it is hard to guarantee either the steady operation or any quality products. Since this draw resonance is an industrially important productivity issue as well as an academically interesting stability topic, not only in film casting but also in fiber spinning and film blowing processes, there have been many experimental and theoretical studies on this subject over the past four decades [8–13].

Among those studies on film casting, film blowing processes, there have been notable efforts to stabilize the processes by manipulating process operating conditions such as cooling and coextrusion, and/or the viscoelastic characteristics of the input polymer materials. Two methods are the most salient and successful: the draw resonance eliminator and the encapsulation die. The ingenious device called draw resonance eliminator developed by Union Carbide for film casting [14] employs a maximum cooling onto the film surface to result in significantly increased process productivity with a much-expanded stability region of the process conditions.

Equally ingenious is the encapsulation die which successfully stabilizes the extrusion coating of low-melt-strength resins like high-density polyethylene (HDPE) and LLDPE by co-extruding small amounts of high-melt-strength resins like low-density polyethylene (LDPE) side-by-side, i.e. encapsulating HDPE in the center [15]. It exhibits minimal neck-in while offering excellent draw properties and web stability for uniform coating. In this study, we explain why such encapsulation extrusion method is so successful in stabilizing the draw resonance in extrusion coating process. The simulation method of the present study can also be employed to help develop strategies for stabilizing other film casting, film coating, and film blowing processes.

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2. Formulation of the model

As shown in Fig. 1, the encapsulation coating employs the coextrusion of a high-melt-strength material attached to the outer edges of a core low-melt-strength material in a flat die. For the simulation of the transient behavior of the film in this film casting system with encapsulation dies, and in order to portray draw resonance instability and neck-in phenomenon, we have employed in the present study an isothermal one-dimensional varying film width model. This model is based on two excellent articles in this field, i.e. the one by Pis-Lopez and Co [16] for multilayer film casting and the other by Silagy et al. [3] with the free film surface edge conditions which enable the prediction of the neck-in phenomenon. A Phan-Thien–Tanner (PTT) constitutive equation [17,18] is adopted as a viscoelastic fluid model, known for its robustness and accuracy in describing extensional deformation processes [19]. The model has the following form:

Continuity equation:

$$\frac{\partial(ew_1)}{\partial t}+\frac{\partial(ew_1v)}{\partial x}=0, \tag{1}$$

$$\frac{\partial[e(w_2-w_1)]}{\partial t}+\frac{\partial[e(w_2-w_1)v]}{\partial x}=0, \tag{2}$$

where $e=\bar{e}/\bar{e}_0$, $w_1=\bar{w}_1/\bar{w}_{10}$, $w_2=\bar{w}_2/\bar{w}_{10}$, $v=\bar{v}/\bar{v}_0$, $t=\bar{t}\bar{v}_0/L$, $x=\bar{x}/L$.

Equation of motion:

$$F=\sigma_{1xx}ew_1+V_{\mathrm{r}}\sigma_{2xx}e(w_2-w_1), \tag{3}$$

where $F=\bar{F}L/4\eta_1\bar{e}_0\bar{w}_{10}\bar{v}_0$, $\sigma_{1ij}=\bar{\sigma}_{1ij}L/\eta_1\bar{v}_0$, $\sigma_{2ij}=\bar{\sigma}_{2ij}L/\eta_2\bar{v}_0$, $V_{\mathrm{r}}=\eta_2/\eta_1$.

Constitutive equations (PTT fluids):

$$K_1\boldsymbol{\tau}_1+De_1\left[\frac{\partial\boldsymbol{\tau}_1}{\partial t}+\boldsymbol{v}\cdot\nabla\boldsymbol{\tau}_1-\boldsymbol{L}_1\cdot\boldsymbol{\tau}_1-\boldsymbol{\tau}_1\cdot\boldsymbol{L}_1^{\mathrm{T}}\right]=2\boldsymbol{D}_1, \tag{4}$$

$$K_2\boldsymbol{\tau}_2+De_2\left[\frac{\partial\boldsymbol{\tau}_2}{\partial t}+\boldsymbol{v}\cdot\nabla\boldsymbol{\tau}_2-\boldsymbol{L}_2\cdot\boldsymbol{\tau}_2-\boldsymbol{\tau}_2\cdot\boldsymbol{L}_2^{\mathrm{T}}\right]=2\boldsymbol{D}_2, \tag{5}$$

where

$$K_1=\exp\left[\varepsilon_1De_1\mathrm{tr}(\boldsymbol{\tau}_1)\right],\ K_2=\exp\left[\varepsilon_2De_2\mathrm{tr}(\boldsymbol{\tau}_2)\right]$$

$$\boldsymbol{L}_1=\nabla\boldsymbol{v}-\xi_1\boldsymbol{D}_1,\quad \boldsymbol{L}_2=\nabla\boldsymbol{v}-\xi_2\boldsymbol{D}_2$$

$$\tau_{1ij}=\bar{\tau}_{1ij}L/\eta_1\bar{v}_0,\quad \tau_{2ij}=\bar{\tau}_{2ij}L/\eta_2\bar{v}_0,$$
$$De_1=\lambda_1\bar{v}_0/L,\quad De_2=\lambda_2\bar{v}_0/L.$$

Film edge conditions:

$$\sigma_{2xx}\left(\frac{\partial w_2}{\partial x}\right)^2=A_{\mathrm{r}}^2\sigma_{2yy},\quad \sigma_{1zz}=\sigma_{2zz}=0, \tag{6}$$

where $A_{\mathrm{r}}=L/\bar{w}_{10}$.

Boundary conditions:

$$\begin{aligned}
t=0:\quad & e=(e)_{\mathrm{s.s.}},\ w_1=(w_1)_{\mathrm{s.s.}},\ w_2=(w_2)_{\mathrm{s.s.}},\ v=(v)_{\mathrm{s.s.}},\ \tau_{1xx}=(\tau_{1xx})_{\mathrm{s.s.}},\\
& \sigma_{1xx}=(\sigma_{1xx})_{\mathrm{s.s.}},\ \sigma_{1yy}=(\sigma_{1yy})_{\mathrm{s.s.}},\ \tau_{2xx}=(\tau_{2xx})_{\mathrm{s.s.}},\ \sigma_{xx2}=(\sigma_{2xx})_{\mathrm{s.s.}},\ \sigma_{2yy}=(\sigma_{2yy})_{\mathrm{s.s.}}\\
t>0:\quad & e=e_0=1,\ w_1=w_{10}=1,\ w_2=w_{20}=1+q,\ v=v_0=1, \quad \text{at}\ x=0\\
& v=v_{\mathrm{L}}=r(1+\varepsilon^*), \quad \text{at}\ x=1
\end{aligned} \tag{7}$$

where over bars mean dimensional variables; subscripts $i=1$ and $i=2$ represent conditions for core film and outer film, respectively; e is the dimensionless film thickness; w_i is the dimensionless film widths; v is the dimensionless velocity; x is the dimensionless distance; L is the distance between die exit and chill roll; F is the dimensionless axial tension, $\boldsymbol{\sigma}_i$ is the dimensionless total stresses; η_i is the zero-shear viscosities; V_{r} is the viscosity ratio; $\boldsymbol{\tau}_i$ is the dimensionless deviatoric stresses; $\boldsymbol{D}_i$ is the deformation tensors; t is the dimensionless time; λ_i is the material relaxation times; De_i is the Deborah numbers; ε_i, ξ_i are the PTT model parameters; A_{r} is the aspect ratio; r is the film drawdown ratio, i.e. the ratio of film velocity at die exit and at chill roll; q is the relative amount of outer film to core film, ε^* is a constant representing the extent of initial disturbances at the take-up.

Subscripts 0, L, and s.s. denote values at die exit, take-up, and steady state conditions, respectively. The relations between $\boldsymbol{\sigma}$ and $\boldsymbol{\tau}$ are explained in Appendix A along with the edge conditions of Eq. (6).

The following assumptions have been incorporated in this one-dimensional model to focus on the extensional deformation, which constitutes the dominant dynamics in film casting process: The thin film approximation simplifies this system to one-dimensional model with varying film width. The edge beads and thermal effects have been neglected in this 1-D model (the effect of edge beads on the film casting dynamics was elucidated in Debbaut et al. [12]). The secondary forces on the film such as gravity, inertia, air

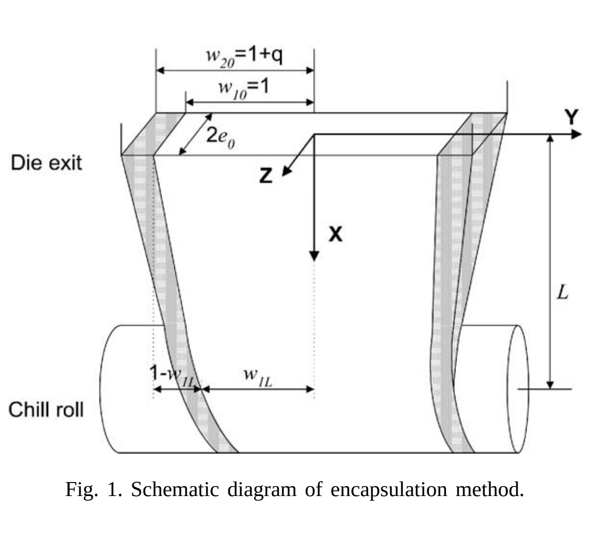

Fig. 1. Schematic diagram of encapsulation method.

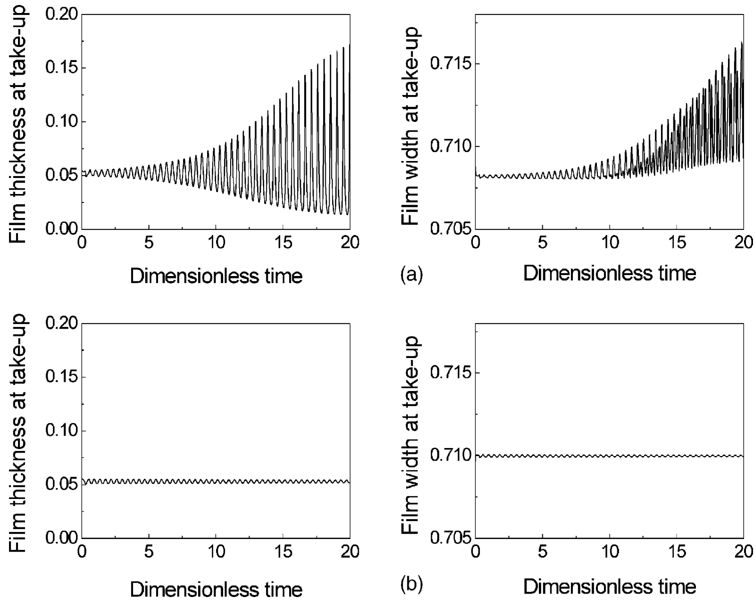

Fig. 2. Transient response of cast film processes (a) an extension thinning material ($r = 27$, $A_r = 0.5$, $e_1 = 0.015$, $x_1 = 0.7$, $De_1 = 0.005$, and $q = 0$) and (b) an extension thickening material ($r = 27$, $A_r = 0.5$, $e_1 = 0.015$, $x_1 = 0.1$, $De_1 = 0.005$, and $q = 0$).

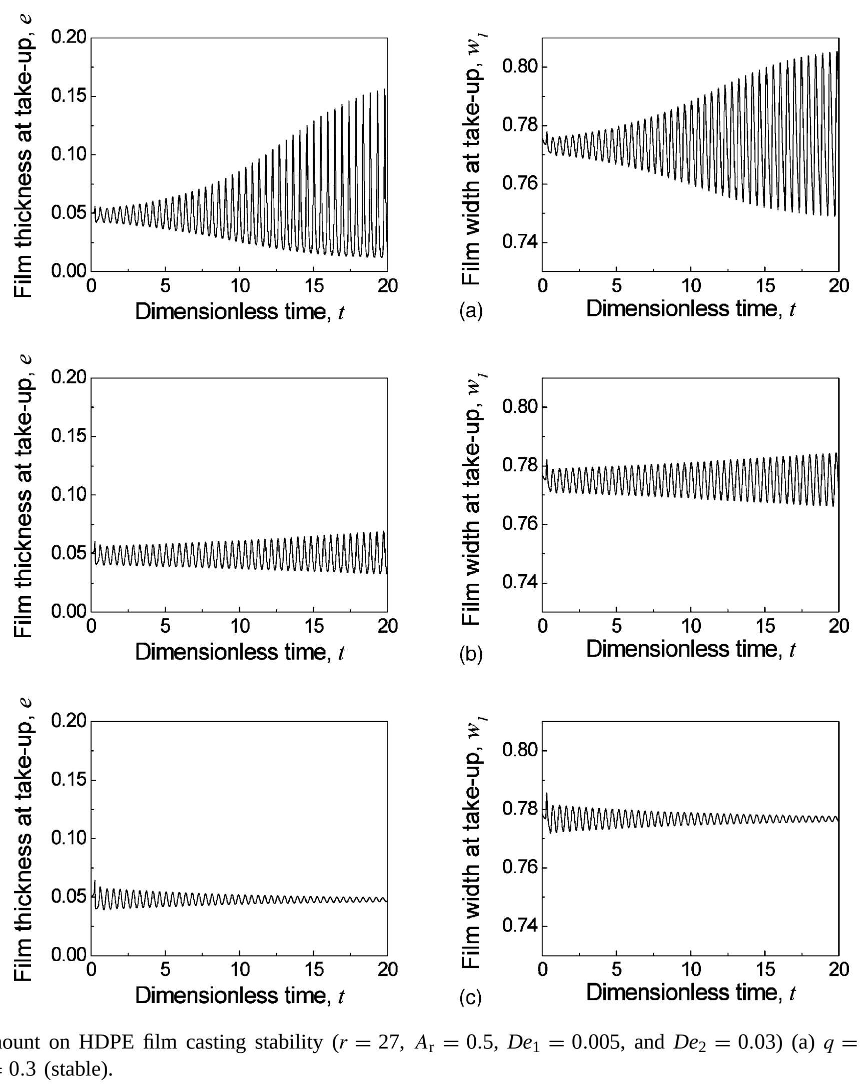

Fig. 3. Effect of LDPE amount on HDPE film casting stability ($r = 27$, $A_r = 0.5$, $De_1 = 0.005$, and $De_2 = 0.03$) (a) $q = 0.1$ (unstable), (b) $q = 0.2$ (less unstable), and (c) $q = 0.3$ (stable).

drag, and surface tension are also neglected because they are usually small in film casting process.

In this 1-D model, the axial distance, x, is the only space coordinate of the system with film stresses, thus, not varying in the y-coordinate within each polymer phase. As a consequence, the boundary conditions at the interface between the core and the outer films do not enter the model because the interface boundary conditions become irrelevant in 1-D models. The interface boundary conditions, of course, should be included when a more accurate 2-D model is employed to solve the same encapsulation extrusion problem, which is presently under way in the authors' laboratory with the results to be reported elsewhere in the future.

Transient solutions of the above governing Eqs. (1)–(7) have been obtained using a first-order upwind implicit finite difference method to avoid numerical instability problems caused by the fluid viscoelasticity, particularly at high Deborah numbers. It also has been found that the x–t grid of $2000 \times 10,000$ mesh points guarantees acceptable accuracy for numerical results in typical viscoelastic cases [7,11,20,21].

3. Discussion

In order to examine the effect of the encapsulation extrusion method on the stability of film casting process, numerical simulation has been performed with different outer LDPE film widths in conjunction with different fluid viscoelasticity values. The transient response of the film width and thickness to system disturbances has been obtained solving the above governing equations when disturbances (e.g. a 5% step-change) were introduced in the film take-up velocity at the chill roll. The stability of the process then

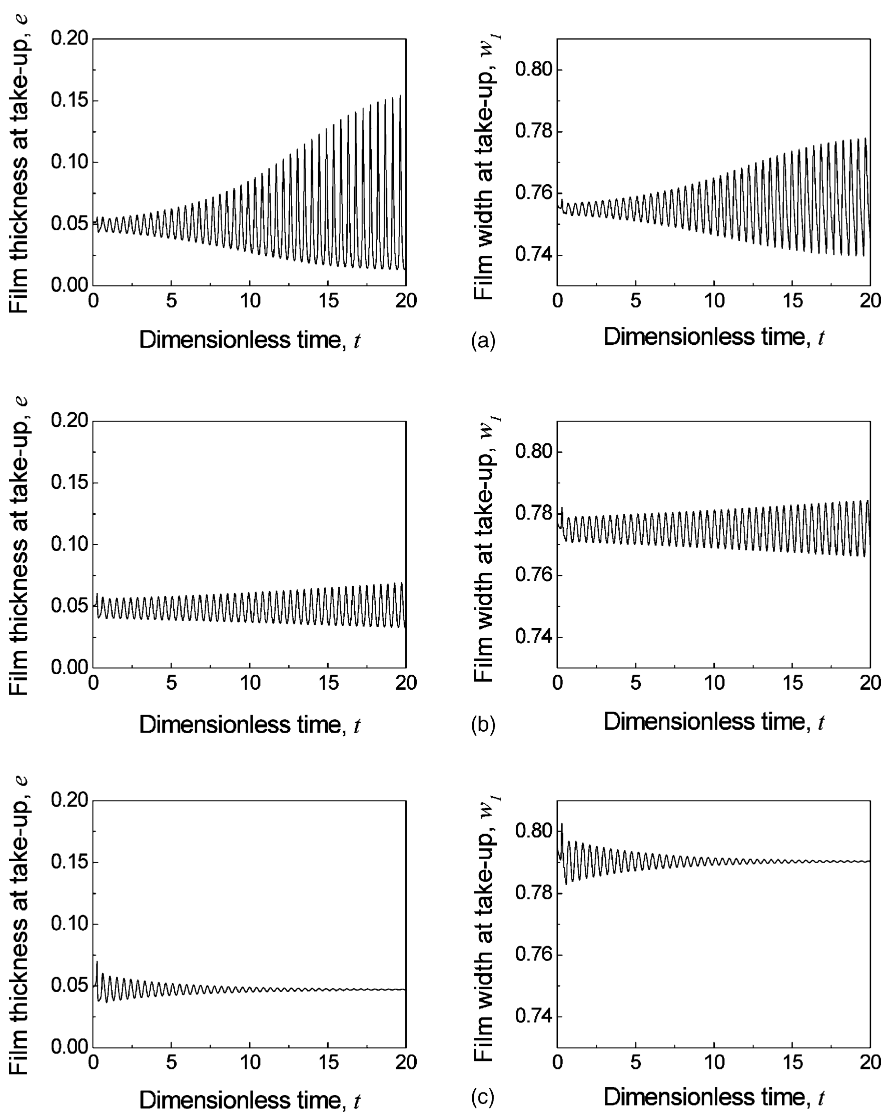

Fig. 4. Effect of viscoelasticity of LDPE on HDPE film casting ($r = 27$, $A_r = 0.5$, $De_1 = 0.005$, and $q = 0.2$) (a) $De_2 = 0.02$ (unstable), (b) $De_2 = 0.03$ (less unstable), and (c) $De_2 = 0.04$ (stable).

can be determined by observing the temporal characteristics of the film width and thickness as time evolves. The values of PTT model parameters for both core and outer film materials were adopted from the literature [18,22].

First, Fig. 2 shows that pure HDPE film casting is unstable while pure LDPE film casting at the same process conditions is stable. This is because the extension-thinning HDPE is generally less stable than extension-thickening LDPE in extensional deformation processes as reported in the literature [20–24].

Now, as shown in Fig. 3, the simulation results reveal that as the amount of LDPE attached to both outer edges of the core HDPE film increases, the process becomes more stabilized. As the value of q, the amount of outer film relative to that of the core film, increases from 0.1 to 0.3, both film thickness and film width exhibit progressively more stable behavior.

Moreover, as exhibited in the film width response, the film width reduction at the take-up, the so-called "neck-in phenomenon", becomes less severe as the LDPE amount increases as shown in Fig. 3. In other words, the two undesirable phenomena occurring in film casting, i.e. draw resonance and neck-in phenomenon, can both be stabilized by this encapsulation co-extrusion method.

What we have demonstrated in Fig. 3 is that our model of Eqs. (1)–(7) has produced the same results as experimentally observed, thus, successfully explaining why the encapsulation extrusion of HDPE with LDPE can stabilize the process. The key to this stabilization is the axial tension in Eq. (3) which is substantially increased by the addition of the high melt-strength material LDPE, albeit its small amount. This is possible due to the fact that the axial stress generated by LDPE is much higher than that of HDPE, which is obviously responsible for the different stability behavior of the two materials as exhibited in Fig. 2.

Since the tension increasing effect by the outer LDPE film is responsible for the stabilization of core HDPE film casting, it is obvious that the reverse case, i.e. the core LDPE with outer HDPE, which would have resulted in tension reduction, does not work. Understandably, there have not been any reports on these reverse cases.

Since the amount of the outer LDPE should be minimized to reduce the cost of stabilizing HDPE extrusion coating, different viscoelasticity values of the LDPE were used in other simulation runs. As shown in Fig. 4, the stabilizing effect of LDPE increases with its increasing Deborah number. This is because the tension provided by the outer LDPE to the system increases with its Deborah number, which is a well known fact for extensional thickening materials like LDPE in extensional deformation processes such as fiber spinning, film casting and film blowing [11,20–24].

One comment is in order now as to why some polymer extrusion coating has been found not to be enhanced by encapsulating extrusion: The case of PET extrusion coating not stabilized by coextruding LDPE at the outer edge [15]. The explanation for this industrially important finding is rather simple: The rather high processing temperatures of PET of 280 °C makes the viscosity of LDPE so low at that temperature that the additional tension provided by LDPE does not make much difference in stabilizing the extrusion of PET film. Furthermore, when the viscosity of LDPE is small at high temperatures, the stability of its own extrusion deteriorates, too. The combination of these two effects renders the encapsulation extrusion of PET by LDPE useless. Fig. 5 shows the simulation results of this system.

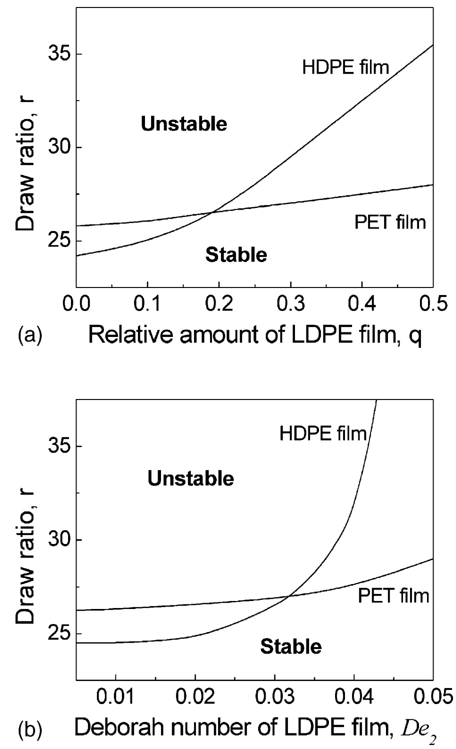

Fig. 5. Stability diagram of HDPE ($e_1 = 0.015$, $x_1 = 0.7$, $A_r = 0.5$, $De_1 = 0.005$, $V_r = 1.0$ at 180 °C) and PET ($e_1 = 0.0$, $x_1 = 0.0$, $A_r = 0.5$, $De_1 = 0.0005$, $V_r = 0.5$ at 280 °C) film casting with outer film of LDPE. (a) Effect of LDPE amount (q) when $De_2 = 0.03$, and (b) effect of fluid viscoelasticity of LDPE (De_2) when $q = 0.2$.

Finally, one more remark regarding the 1-D and 2-D models in connection with the neck-in phenomenon (film width reduction) is in order. Although 2-D models produce more accurate results on this neck-in (especially in the casting of viscoelastic fluids) as demonstrated in this study, 1-D models are also capable of doing the job reasonably well. In other words, 1-D models suffice to explain the basic physical reasons behind the success of the encapsulation extrusion method, i.e. the objective of this paper. As mentioned before, an effort is currently under way in the authors' laboratory to employ a 2-D model in solving the same encapsulation extrusion coating process.

4. Conclusions

A nonlinear stability analysis of the film casting process with an encapsulation extrusion method is carried out using an isothermal one-dimensional varying width model in order to demonstrate why the encapsulation extrusion coating process is so successful industrially for low-melt-strength polymers like HDPE when coextruding high-melt-strength materials like LDPE side by side. The success is due to the fact that the total axial stress, which is known to be responsible for stabilization of extensional deformation processes such as fiber spinning, film casting and film blowing [20–24], increases dramatically by the addition of the small amounts of the outer-attached LDPE film. The simulation, thus, theoretically explains why and how the encapsulation extrusion of HDPE with LDPE outer-attached stabilizes the process. It also exemplifies the importance of film width control in the industrially important paper coating process.

The fact that the extrusion coating of PET is, however, not stabilized by the same encapsulation with LDPE, is also easily explained by the simulation model of the present study. That is, at the higher extrusion temperatures of PET, the axial stress-increasing-effect by LDPE is to make much difference, meaning that we need to find materials other than LDPE with higher extensional viscosity for stabilizing PET film coating.

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Appendix A

The total normal stress on the free surfaces of the film side edges is set to zero when surface tension is negligible [3].

$$\boldsymbol{\sigma}_2 \cdot \boldsymbol{n} = \mathbf{0}, \tag{A.1}$$

$$\begin{pmatrix} \sigma_{2xx} & \sigma_{2xy} & \sigma_{2xz} \\ \sigma_{2xy} & \sigma_{2yy} & \sigma_{2yz} \\ \sigma_{2xz} & \sigma_{2yz} & \sigma_{2zz} \end{pmatrix} \begin{pmatrix} \sin\alpha \\ \cos\alpha \\ 0 \end{pmatrix} = \begin{pmatrix} \tau_{2xx}-P & \tau_{2xy} & \tau_{2xz} \\ \tau_{2xy} & \tau_{2yy}-P & \tau_{2yz} \\ \tau_{2xz} & \tau_{2yz} & \tau_{2zz}-P \end{pmatrix} \begin{pmatrix} \sin\alpha \\ \cos\alpha \\ 0 \end{pmatrix} = \begin{pmatrix} 0 \\ 0 \\ 0 \end{pmatrix}, \tag{A.2}$$

where α is the angle between normal direction at the free surface and y-axis. Then,

$$\tau_{2xy} = -\frac{\sin\alpha}{\cos\alpha}(\tau_{2xx}-P) = -(\tau_{2xx}-P)\tan\alpha = A_{\mathrm{r}}\left(\frac{\partial w_2}{\partial x}\right)(\tau_{2xx}-P), \tag{A.3}$$

$$\tau_{2yy}-P = -\frac{\sin\alpha}{\cos\alpha}\tau_{2xy} = -\tau_{2xy}\tan\alpha = A_{\mathrm{r}}\left(\frac{\partial w_2}{\partial x}\right)\tau_{2xy}. \tag{A.4}$$

The membrane approximation of $\sigma_{2zz} = \tau_{2zz} - P = 0$ leads to

$$(\tau_{2xx}-\tau_{2zz})\left(\frac{\partial w_2}{\partial x}\right)^2 = A_{\mathrm{r}}^2(\tau_{2yy}-\tau_{2zz}). \tag{A.5}$$

or

$$\sigma_{2xx}\left(\frac{\partial w_2}{\partial x}\right)^2 = A_{\mathrm{r}}^2\sigma_{2yy}. \tag{A.6}$$

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