

Nonlinear dynamics and stability of film casting process

Joo Sung Lee and Jae Chun Hyun

Department of Chemical Engineering, Applied Rheology Center
Korea University, Seoul 136-701, Korea

(Received November 15, 2001)

Abstract

As part of continuing efforts to investigate nonlinear dynamics and stability of film casting process, our earlier results obtained by Lee *et al.* (2001b) have been extended in the present study to cover the film casting of both extension thickening and extension thinning fluids. The same instability mechanism and draw resonance criterion previously derived have been found valid here, and a rather complex dynamic behavior of film width in contrast to that of film thickness has also been confirmed. The effect of fluid viscoelasticity on draw resonance, however, exhibits opposite results depending on whether the fluid is extension thickening or thinning, i.e., it stabilizes film casting in the former while destabilizing in the latter. The encapsulation extrusion method which recently has been successfully employed to stabilize industrially important paper coating process, has been theoretically explained in the present study as to why such stabilization is possible.

Keywords : draw resonance, encapsulation die, extension thickening, extension thinning, film casting, film thickness, film width, viscoelasticity

1. Introduction

Polymer melts are extruded in film casting processes through a slit die either in single or multiple layers to form films for various end uses such as tapes and packaging films (Agassant *et al.*, 1991; Kanai and Campbell, 1999). The molten film cast out of the extrusion die is stretched and cooled in the air before reaching the chill roll on which the film moves with a speed faster than that at the die exit, resulting in a larger-than-unity drawdown ratio. When this drawdown ratio is increased beyond a critical value, the film casting process can become unstable, i.e., instability called draw resonance occurs, in spite of constant extrusion and take-up speeds (Luchesi *et al.*, 1985; Anturkar and Co, 1988; Iyengar and Co, 1996; Silagy *et al.*, 1996; Jung *et al.*, 1999). Since this draw resonance is an industrially important productivity issue as well as an academically interesting stability topic in not only in film casting but also in other extension deformation processes like fiber spinning and film blowing, there have been many experimental and theoretical studies on this subject in last four decades (Pearson and Matovich, 1969; Fisher and Denn, 1976; Jung and Hyun, 1999; Yoon and Park, 2000, Lee *et al.*, 2001a).

In the present simulation study on film casting, we have

chosen Agassant's model (Silagy *et al.*, 1996) which has proven very robust in simulating the steady state and transient dynamics of film casting along with a Phan-Thien-Tanner constitutive model known for its capability of describing both extension-thickening and extension-thinning fluids portraying low-density and high-density polyethylenes frequently found in various polymer processing operations.

Just as in the previous article by Lee *et al.* (2001b), the kinematic waves approach which had been developed in spinning process (Kim *et al.*, 1996; Hyun, 1999; Jung *et al.*, 2000) has been adopted to analyze the draw resonance in film casting for both extension thickening and thinning fluids. We also study the nonlinear dynamics of film casting in draw resonance with particular focus on the differences in the transient behavior between the film thickness and the film width. The proper control of film width is of great importance industrially as that of film thickness, as evidenced in the extrusion coating of polymer films on paper substrates. Recently, an ingenious coating method called encapsulation extrusion (Frey *et al.*, 2001) demonstrates that film width fluctuations can be successfully stabilized employing the new co-extrusion method. We will explain why this is possible by theoretically deriving simulation results using the same film casting model of this study.

2. Model formulation

As in the previous study by Lee *et al.* (2001b), for the

*Corresponding author: jchyun@grtrkr.korea.ac.kr
© 2001 by The Korean Society of Rheology

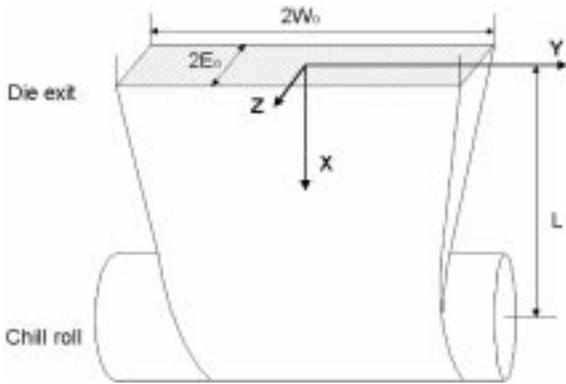


Fig. 1. Schematic of film casting process.

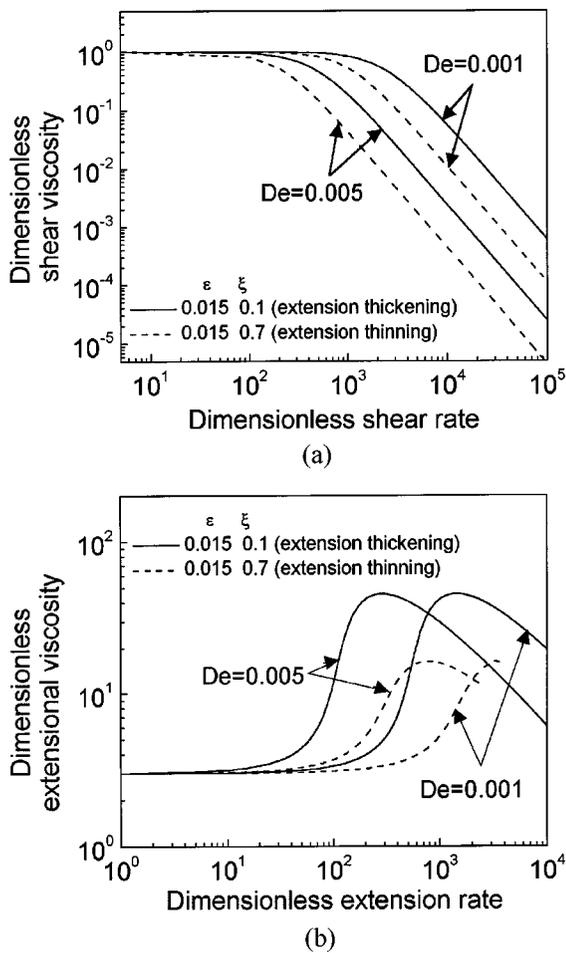


Fig. 2. Response of PTT model for extension thickening (\$\epsilon=0.015, \xi=0.1\$) and thinning fluids (\$\epsilon=0.015, \xi=0.7\$) for (a) simple shear flow and (b) simple extensional flow.

film casting process depicted in Fig. 1, we use in the present study an isothermal one-dimensional varying film width model which Silagy *et al.* (1996) developed and successfully used to obtain excellent results. A Phan-Thien-Tanner constitutive equation (Phan-Thien and Tanner, 1977; Phan-Thien, 1978) is employed as a viscoelastic

fluid model in the present study. PTT model is known for its robustness and accuracy in describing extensional deformation processes (Khan and Larson, 1987) for both extension-thickening and thinning fluids. An example of shear and extensional behavior of PTT model with different values of Deborah number for both fluids are shown in Fig. 2.

$$\text{Continuity equation: } \frac{\partial(e w)}{\partial t} + \frac{\partial(e w v)}{\partial x} = 0 \quad (1)$$

where, \$e = E/E_0\$, \$w = W/W_0\$, \$v = V/V_0\$, \$t = t'V_0/L\$, \$x = X/L\$

$$\text{Equation of motion: } \frac{\partial F}{\partial x} = \frac{\partial(\sigma_{xx} e w)}{\partial x} = \frac{\partial[(\tau_{xx} - \tau_{zz}) e w]}{\partial x} = 0 \quad (2)$$

where, \$\sigma_{ij} = \bar{\sigma}_{ij} L / \eta_0 V_0\$, \$\tau_{ij} = \bar{\tau}_{ij} L / \eta_0 V_0\$,

Constitutive equations (PTT fluids):

$$K \tau_{xx} + De \left[\frac{\partial \tau_{xx}}{\partial t} + v \frac{\partial \tau_{xx}}{\partial x} - 2(1 - \xi) \tau_{xx} \frac{\partial v}{\partial x} \right] = 2 \frac{\partial v}{\partial x} \quad (3)$$

$$K \sigma_{xx} + De \left[\frac{\partial \sigma_{xx}}{\partial t} + v \frac{\partial \sigma_{xx}}{\partial x} - 2(1 - \xi) \tau_{xx} \left\{ 2 \frac{\partial v}{\partial x} + \frac{1}{w} \frac{\partial w}{\partial t} + \frac{v}{w} \frac{\partial w}{\partial x} \right\} \right] + 2De(1 - \xi) \sigma_{xx} \left\{ \frac{\partial v}{\partial x} + \frac{1}{w} \frac{\partial w}{\partial t} + \frac{v}{w} \frac{\partial w}{\partial x} \right\} = 2 \left[2 \frac{\partial v}{\partial x} + \frac{1}{w} \frac{\partial w}{\partial t} + \frac{v}{w} \frac{\partial w}{\partial x} \right] \quad (4)$$

$$K \sigma_{yy} + De \left[\frac{\partial \sigma_{yy}}{\partial t} + v \frac{\partial \sigma_{yy}}{\partial x} + 2(1 - \xi) (\sigma_{xx} - \tau_{xx}) \left\{ \frac{\partial v}{\partial x} + \frac{2}{w} \frac{\partial w}{\partial t} + \frac{2v}{w} \frac{\partial w}{\partial x} \right\} \right] - 2De(1 - \xi) \sigma_{yy} \left\{ \frac{1}{w} \frac{\partial w}{\partial t} + \frac{v}{w} \frac{\partial w}{\partial x} \right\} = 2 \left[\frac{\partial v}{\partial x} + \frac{2}{w} \frac{\partial w}{\partial t} + \frac{2v}{w} \frac{\partial w}{\partial x} \right] \quad (5)$$

where, \$K = \exp(\epsilon De \text{trace}(\underline{\tau}))\$, \$De = \lambda_0 V_0 / L\$

$$\text{Film edge conditions: } \sigma_{xx} \left(\frac{\partial w}{\partial x} \right)^2 = A_r^2 \sigma_{yy}, \quad \sigma_{zz} = 0 \quad (6)$$

where, \$A_r = L/W_0\$

Boundary conditions:

$$\begin{aligned} t=0: & e=e_s, w=w_s, v=v_s, \tau=\tau_{s,s}, \sigma=\sigma_{s,s}, \sigma=\sigma_{s,s} \\ t>0: & e=e_0=1, w=w_0=1, v=v_0=1 \text{ at } x=0 \\ & v=v_L=r(1+\epsilon^*) \quad \text{at } L \end{aligned} \quad (7)$$

where, \$e\$ = dimensionless film thickness, \$w\$ = dimensionless film width, \$v\$ = dimensionless film velocity, \$x\$ = dimensionless distance, \$L\$ = distance between die exit and chill roll, \$F\$ = tension on the film, \$\sigma_{ij}\$ = dimensionless total stress in \$ij\$-direction, \$\tau_{ij}\$ = dimensionless stress in \$ij\$-direction, \$t'\$ = time, \$t\$ = dimensionless time, \$\lambda\$ = material relaxation time, \$De\$ = Deborah number, \$\epsilon, \xi\$ = PTT model parameters, \$A_r\$ = aspect ratio, \$r\$ = drawdown ratio, \$\epsilon^*\$ = a constant representing the initial disturbances at the take-up, and subscripts 0, L, S denote die exit, take-up, and steady state conditions, respectively.

Several assumptions have been incorporated in the model. The edge effects (i.e., edge beads) and thermal effects are neglected because of this isothermal one-dimensional model, and the secondary forces on the film such as gravity, inertia, air drag and surface tension are also neglected because they are usually small in film casting process. Finally, the free surface conditions on the film shown in Eq. (6) are the results of the assumption that normal stress vanishes on the film surfaces. Particularly on the film edge,

$$\underline{\underline{\sigma}} \cdot \underline{\underline{n}} = \begin{pmatrix} \sigma_{xx} & \sigma_{xy} & \sigma_{xz} \\ \sigma_{xy} & \sigma_{yy} & \sigma_{yz} \\ \sigma_{xz} & \sigma_{yz} & \sigma_{zz} \end{pmatrix} \cdot \begin{pmatrix} \sin \alpha \\ \cos \alpha \\ 0 \end{pmatrix} = \begin{pmatrix} 0 \\ 0 \\ 0 \end{pmatrix} \quad (8)$$

where $\underline{\underline{\sigma}}$ = total stress tensor,
 $\underline{\underline{n}}$ = normal vector to the film edge surface,
 α = angle between film edge free surface and x-coordinate,

$$\tan \alpha = -\frac{1}{A_r} \left(\frac{dw}{dx} \right)$$

(The detailed derivation of this result can be found in Agassant *et al.* (1991).)

3. Kinematic waves and draw resonance criterion

Following the same procedure as in Lee *et al.* (2001b), we have obtained the dynamic pictures, as shown in Fig. 3, of several kinematic waves such as the unity throughput waves and the cross-sectional area waves traveling on the film from the die exit to the chill roll and also computed their traveling times. Then it turns out that the same draw resonance criterion holds in the present film casting study of extension thickening and thinning fluids as it did in the previous articles of film casting (Lee *et al.*, 2001b) and fiber spinning (Kim *et al.*, 1996; Hyun, 1999; Jung *et al.*, 2000). (We had originally derived this draw resonance criterion based on the dynamics of fiber spinning process.)

In other words, the same draw resonance equation shown in Eq. (9) holds for the film casting of both extension thickening and thinning fluids.

$$2t_L + T/2 \geq 2\theta_L \quad \text{at } r \leq r_c \quad (9)$$

where, t_L = traveling time of unity throughput waves, T = oscillation period, θ_L = traveling time of cross-sectional area waves, r_c = critical draw down ratio at the onset of draw resonance.

4. Nonlinear dynamics of film thickness and film width

It has been found that since the dynamic behavior of film thickness is almost the same as that of film cross-sectional area, the traveling time of the film thickness is indistinguishable from that of the film cross-sectional area and thus the same criterion equation like Eq. (9) can be obtained if we use the traveling times of film thickness waves in lieu of those of film cross-sectional area waves.

However, the behavior of film width waves doesn't follow the same pattern, i.e., the traveling times of film width don't satisfy Eq. (9). Moreover, as shown below, the film width in film casting exhibits quite an interesting transient behavior in contrast to that of film thickness, which rather plainly behaves in a similar fashion with the fiber cross-sectional area in spinning.

Fig. 4 shows the transient behavior of film thickness at the chill roll of extension thickening fluids having different values of viscoelasticity (Deborah number). (Simulating the conditions for low density polyethylene film extrusion, we have used $\epsilon=0.015$, $\xi=0.1$ for our PTT model adopting from the literature (Phan-Thien, 1978). Deborah number was varied between 0.005 and 0.015 and the drawdown ratio, r , was at 30 and the aspect ratio, $A_r=0.5$). As both temporal pictures and phase plane trajectories of the film thickness clearly display, the stability of film casting pro-

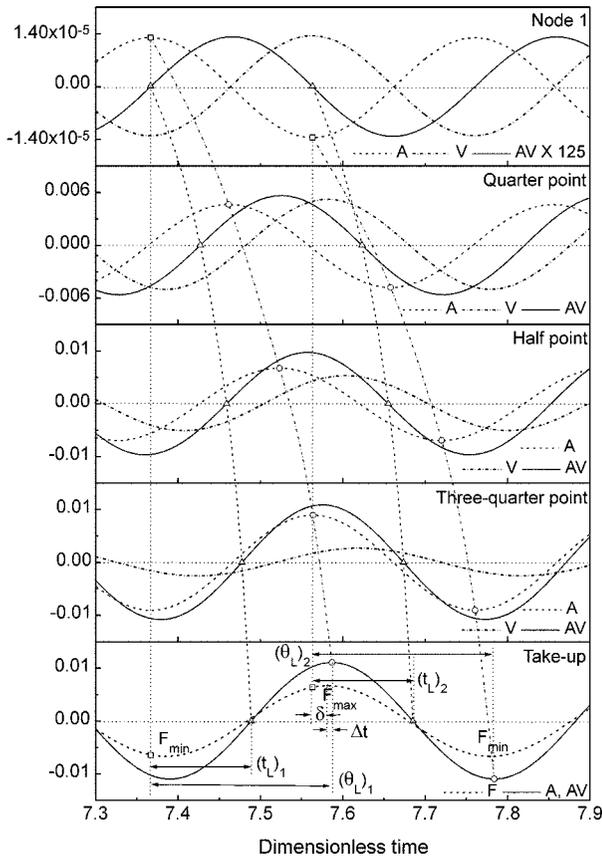


Fig. 3. Transient curves of film variables at five different positions in the casting direction for a viscoelastic PTT fluid ($\epsilon=0.015$, $\xi=0.1$, $De=0.01$) when $A_r=0.2$, and $r=r_c=23.31$: A (cross-sectional area), V (velocity), F (tension force).

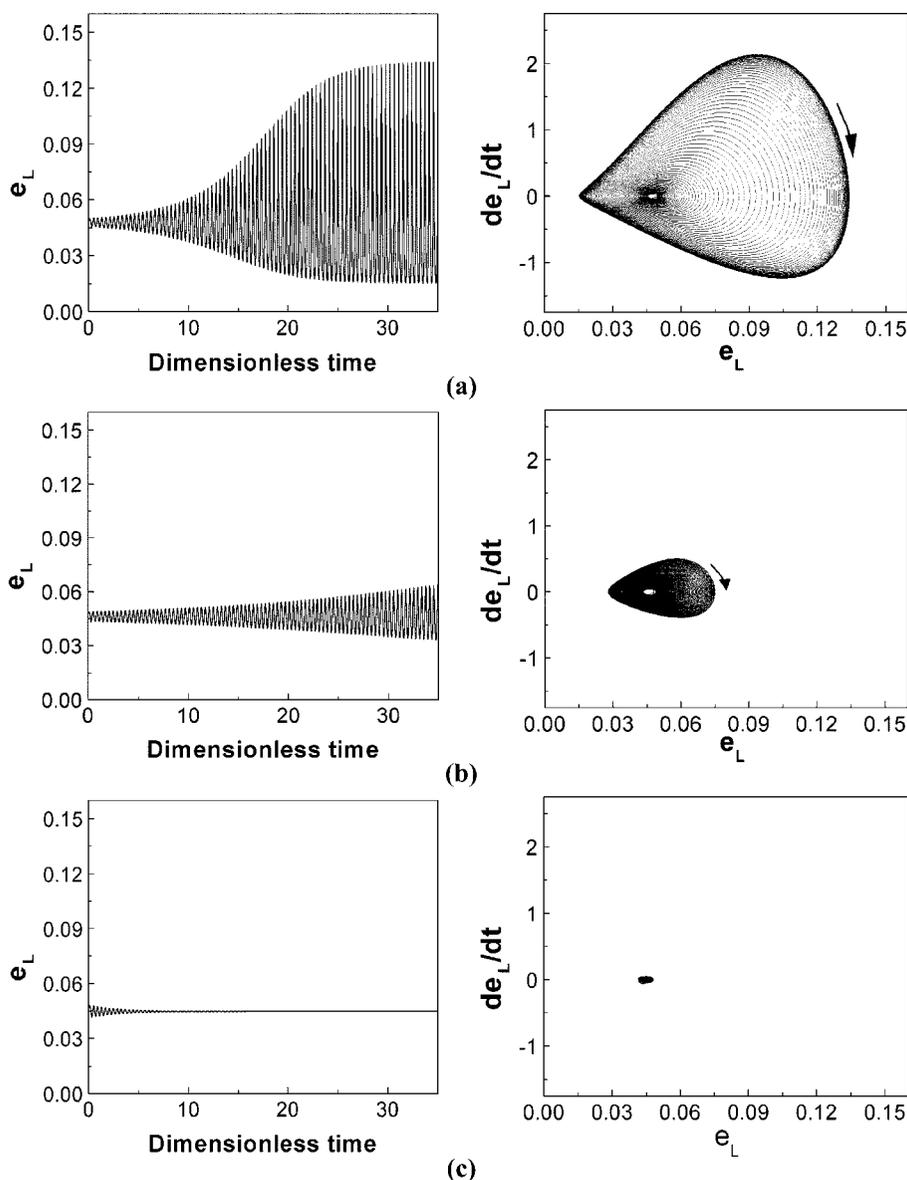


Fig. 4. Transient behavior of film thickness at the take-up of extension thickening fluids ($\varepsilon=0.015$, $\xi=0.1$) having different values of Deborah number when $A_r=0.5$ and $r=30$: (a) $De=0.005$, (b) $De=0.01$, and (c) $De=0.015$.

cess is enhanced by the increasing fluid viscoelasticity.

The transient behavior of film width of the same extension thickening fluids is shown in Fig. 5. While the transient behavior of the film width is quite complex than that of the film thickness, the stability picture is the same, i.e., increasing viscoelasticity stabilizes both the film thickness and film width.

Now we turn to the case of extension thinning fluids. Figs. 6 and 7 show the transient behavior of film thickness and film width of extension thinning fluids having different values of viscoelasticity, respectively. (Simulating the conditions for high density polyethylene film extrusion, we have used $\varepsilon=0.015$, $\xi=0.7$ for our PTT model adopting from the literature (Lee *et al.*, 1995). Deborah number was

varied between 0.001 and 0.007 and the drawdown ratio, r , was at 25 and the aspect ratio, $A_r=0.5$).

Opposite the cases of extension thickening fluids in the above, the stability of film casting of extension thinning fluids worsens with increasing fluid viscoelasticity. This stability picture is the same in both the film thickness (Fig. 6) and the film width (Fig. 7).

A couple of observations with regard to the dynamic behavior of the film width are apparent from the above Figs. 4-7. First, the film width shows a much more complicated behavior than film thickness (i.e., the temporal pictures of film width showing some kinks and wiggles as opposed to rather smooth curves of film thickness), although the magnitude of oscillations of the film width is

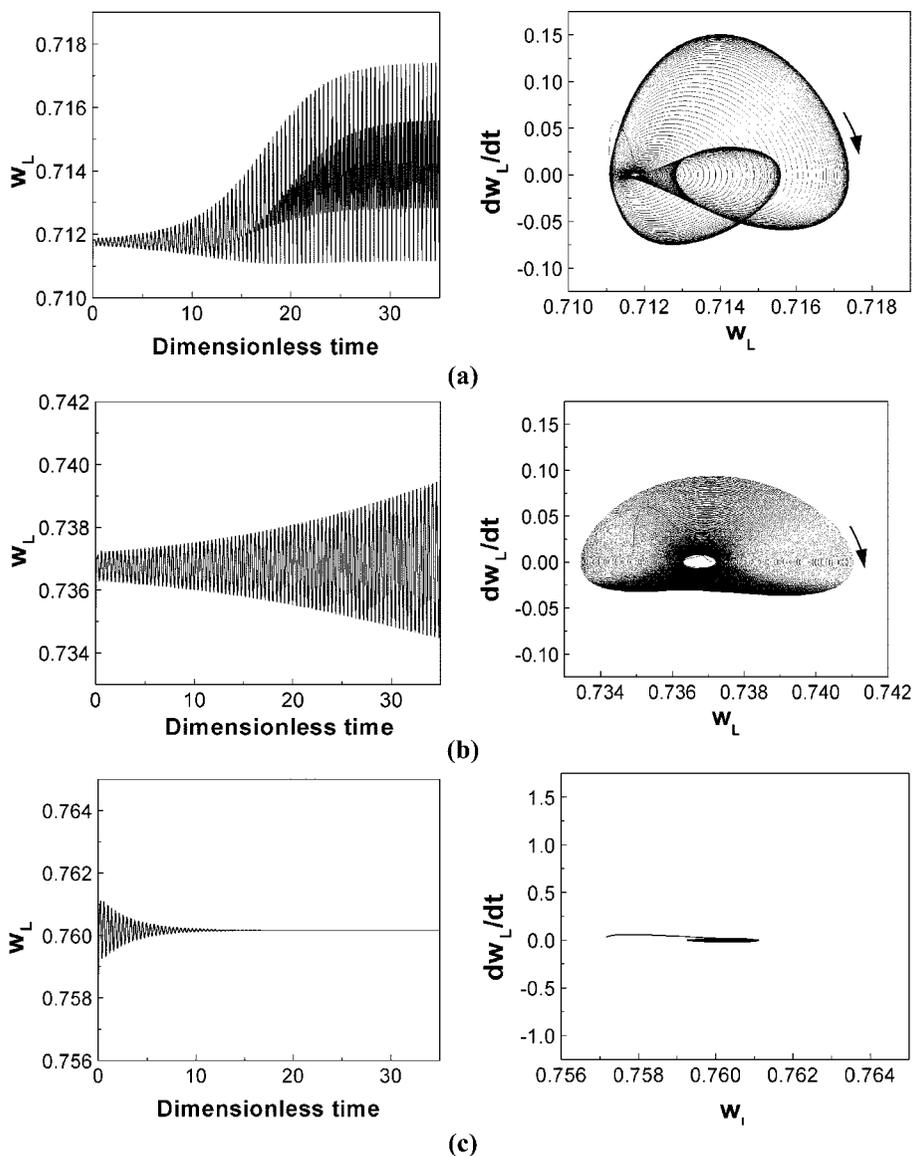


Fig. 5. Transient behavior of film width at the take-up of extension thickening fluids ($\varepsilon=0.015$, $\xi=0.1$) having different values of Deborah number when $A_r=0.5$ and $r=30$: (a) $De=0.005$, (b) $De=0.01$, and (c) $De=0.015$.

smaller than that of the film thickness. Second, the nonlinear dynamic behavior of the film width is sensitive to the viscoelasticity whereas the film thickness does not exhibit much difference in the general pattern of dynamic behavior against the changing viscoelasticity.

The reason why the film width exhibits such a drastically different nonlinear dynamic behavior in draw resonance as compared to the film thickness is due to the fact that we have different edge conditions for the thickness and width. In other words, for the film thickness, vanishing normal stress gives $\sigma_{zz}=0$ on the film surface on x-y plane whereas for the film width vanishing normal stress results in Eq. (6) on the film edges. Because of this difference, it turns out that the dynamics of film width depends on σ_{yy} as well as on σ_{xx} , whereas the film thickness depends mostly on σ_{xx} .

Finally, we proceed to the subject of the encapsulation extrusion method (Frey *et al.*, 2001) which recently has been successfully employed to enhance the stability of paper coating process. As shown in Fig. 8, it has been known that when low melt-strength materials like high-density polyethylene are coated on the paper substrate, more severe stability problem of the film width is encountered than in the case of high melt-strength materials like low-density polyethylene (Hyun and Lee, 2001). In an effort to alleviate this stability problem, more stable LDPE is coextruded at the both outer edges of the less stable HDPE in the core. Industrially, special encapsulation dies (Frey *et al.*, 2001) have been designed and used for this purpose.

In order to simulate this encapsulation extrusion and

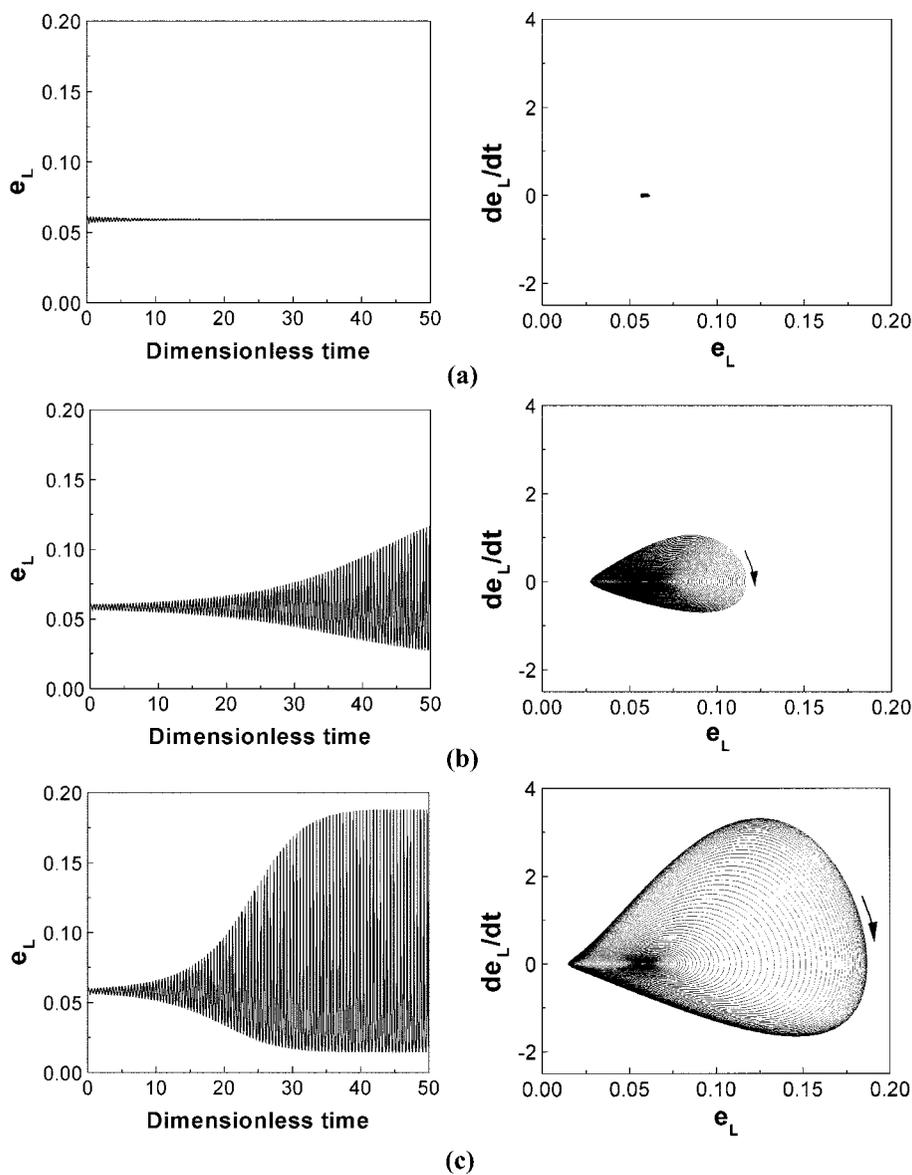


Fig. 6. Transient behavior of film thickness at the take-up of extension thinning fluids ($\varepsilon=0.015$, $\xi=0.7$) having different values of Deborah number when $A_r=0.5$ and $r=25$: (a) $De=0.001$, (b) $De=0.005$, and (c) $De=0.007$.

explain why the coextrusion of LDPE at the edges helps stabilize the film casting of HDPE in the center, we have used the same governing equations of Eqs. (1)-(6) for both the core and edge components. The equation of motion, however, has to be modified to incorporate the shear effects at the interfaces of the two components, and the deformation tensor is also modified likewise to include the shear terms. The results in Fig. 9 clearly demonstrate that the stability of the core component HDPE is enhanced by adding more of LDPE at the both edges, i.e., increasing the value of the parameter q . The stabilizing effect of this encapsulation coextrusion obviously depends on many parameters of the system, i.e., viscosity, elasticity of the two fluids and their ratios between the two components. The

details of this theoretical study explaining the mechanism of the encapsulation extrusion method will be reported elsewhere (Hyun and Lee, 2001).

5. Conclusions

The film casting process has been studied using an isothermal one-dimensional varying width model employing a Phan-Thien-Tanner constitutive equation for both extension thickening and extension thinning fluids. The same mechanism and criterion of draw resonance have been confirmed to hold as in previous articles of film casting and fiber spinning (Kim *et al.*, 1996; Hyun, 1999; Jung *et al.*, 2000; Lee *et al.*, 2001b). Also the film width displays dras-

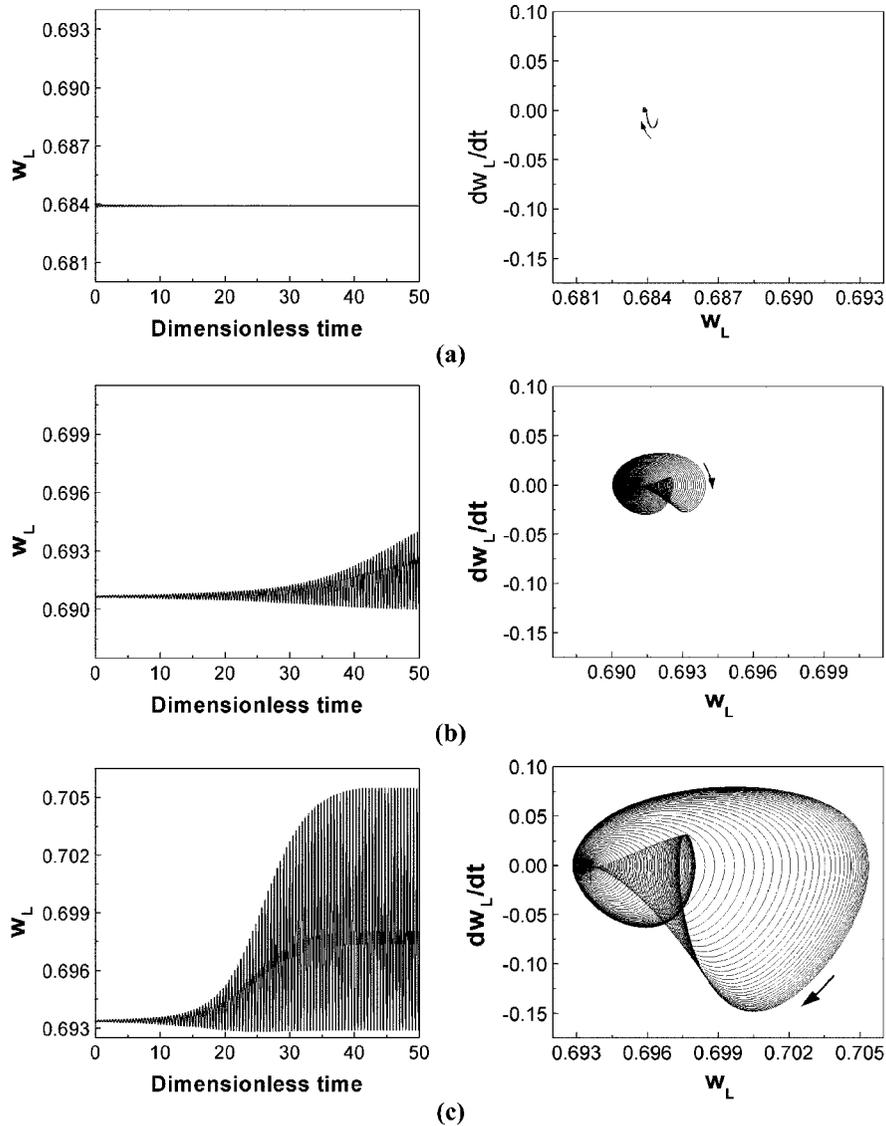


Fig. 7. Transient behavior of film width at the take-up of extension thinning fluids ($\varepsilon=0.015$, $\xi=0.7$) having different values of Deborah number when $A_r=0.5$ and $r=25$: (a) $De=0.001$, (b) $De=0.005$, and (c) $De=0.007$.

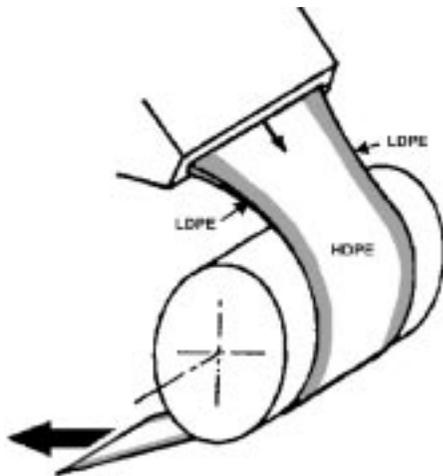


Fig. 8. Schematic of an encapsulation die.

tically different nonlinear dynamic behavior than the film thickness, i.e., the dynamic behavior of film width is more complex and also sensitive to the changing process parameters like fluid viscoelasticity. The stability behavior of extension-thickening and extension-thinning materials in film casting is opposite to each other in that viscoelasticity enhances the stability of the former while it deteriorates for the latter. As far as the stability goes, there is no distinction between the behaviors of film thickness and film width, i.e., their stability responses to fluid viscoelasticity are qualitatively the same.

Encapsulation extrusion method (Frey *et al.*, 2001) exhibiting an ingenious way of stabilizing the film width control in paper extrusion coating process, exemplifies the importance of film width control in industrially important processes. The simulation method of the present study has

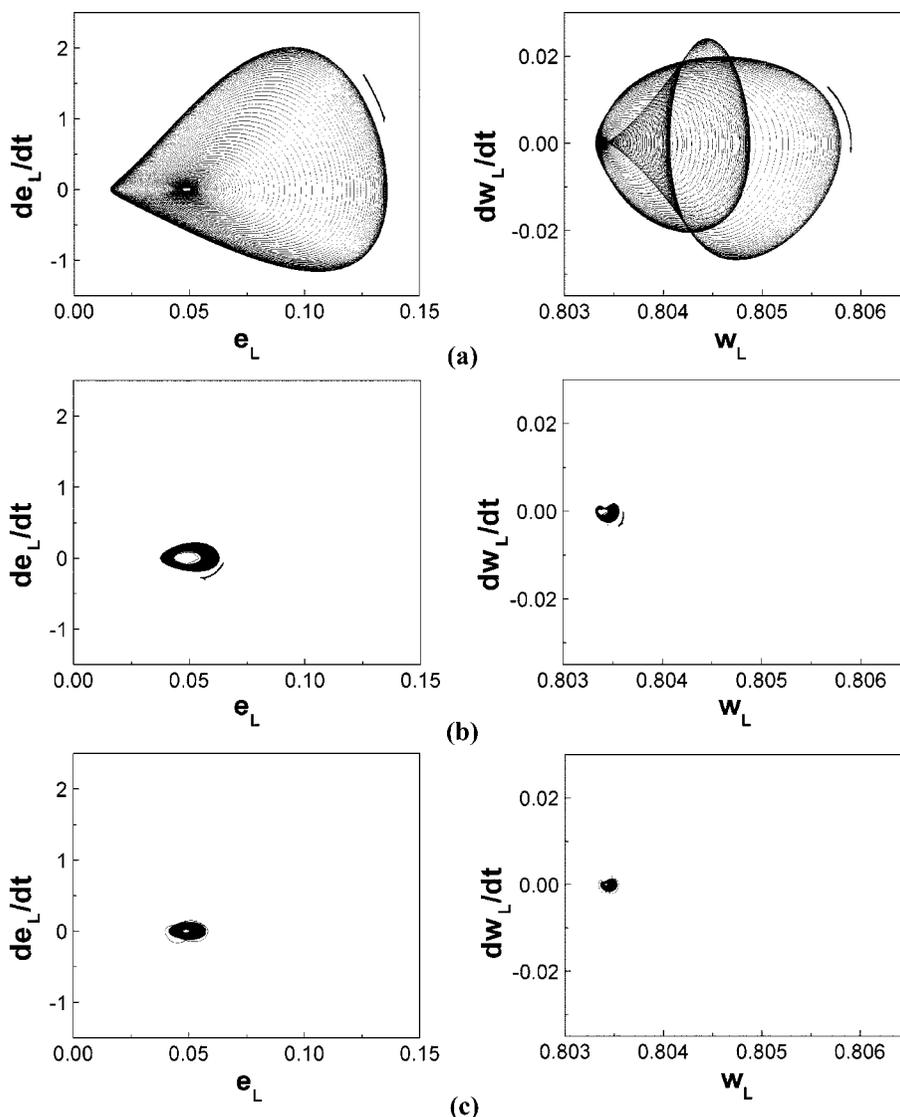


Fig. 9. Effect of encapsulation on the stability when $A_r=0.3$ and $r=30$: (a) $q=0.0$, (b) $q=0.2$, and (c) $q=0.3$ where q is the ratio of the widths of LDPE and HDPE components.

been used to explain why it works by including the shear effects at the interfaces into the model. The details of this simulation will be reported elsewhere.

Acknowledgements

This study was supported by research grants from the Korea Science and Engineering Foundation (KOSEF) through the Applied Rheology Center (ARC), an official KOSEF-created engineering research center (ERC) at Korea University, Seoul, Korea.

References

- Agassant, J.-F., P. Avenas, J.-Ph. Sergent, and P.J. Carreau, 1991, Polymer processing, Hanser Publishers.
- Anturkar, N.R., and A. Co, 1988, Draw resonance in film casting of viscoelastic fluids: a linear stability analysis, *J. Non-Newtonian Fluid Mech.* **28**, 287.
- Fisher, R.J., and M.M. Denn, 1976, A theory of isothermal melt spinning and draw resonance, *AIChE J.* **22**, 236.
- Frey, K.R., G.D. Jerdee, and C.D. Cleaver, 2001, The use of encapsulation dies for processing linear polyolefin resins in extrusion coating, Proc. of ANTEC 2001, Dallas 6-10 May.
- Hyun, J.C., 1999, Draw resonance in polymer processing: a short chronology and a new approach, *Korea-Australia Rheology J.* **11**, 279.
- Hyun, J.C., and J.S. Lee, 2001, Theoretical explanation of process stability enhancement by the encapsulation extrusion coating method, Proc. of Australian-Korean Rheology Conference 2001, Melbourne 20-21 September.
- Iyengar, V.R., and A. Co, 1996, Film casting of a modified Giesekus fluid: stability analysis, *Chem. Eng. Sci.* **51**, 1417.

- Jung, H.W., and J.C. Hyun, 1999, Stability of isothermal spinning of viscoelastic fluid, *Korean J. Chem. Eng.* **16**, 325.
- Jung, H.W., S.M. Choi, and J.C. Hyun, 1999, An approximate method for determining the stability of film-casting process, *AIChE J.* **45**, 1157.
- Jung, H.W., H.-S. Song, and J.C. Hyun, 2000, Draw resonance and kinematic waves in viscoelastic isothermal spinning, *AIChE J.* **46**, 2106.
- Kanai, T., and G.A. Campbell, 1999, Film processing, Hanser Publishers.
- Khan, S.A., and R.G. Larson, 1987, Comparison of simple constitutive equations for polymer melt in shear and biaxial and uniaxial extensions, *J. Rheol.* **31**, 207.
- Kim, B.M., J.C. Hyun, J.S. Oh, and S.J. Lee, 1996, Kinematic waves in the isothermal melt spinning of Newtonian fluids, *AIChE J.* **42**, 3164.
- Lee, J.S., H.W. Jung, S.H. Kim, and J.C. Hyun, 2001a, Effect of fluid viscoelasticity on the draw resonance dynamics of melt spinning, *J. Non-Newtonian Fluid Mech.* **99**, 159.
- Lee, J.S., H.W. Jung, H.-S. Song, K.-Y. Lee, and J.C. Hyun, 2001b, Kinematic waves and draw resonance in film casting process, *J. Non-Newtonian Fluid Mech.* **101**, 43.
- Lee, S., B.M. Kim, and J.C. Hyun, 1995, Dichotomous behavior of polymer melts in isothermal melt spinning, *Korean J. Chem. Eng.* **12**, 345.
- Luchesi, P.J., E.H. Roberts, and S.J. Kurtz, 1985, Reducing draw resonance in LLDPE film resins, *Plast. Eng.*, **41**, 87.
- Pearson, J.R.A., and M.A. Matovich, 1969, Spinning a molten threadline: stability, *I&EC Fund.* **8**, 605.
- Phan-Thien, N., and R.I. Tanner, 1977, A new constitutive equation derived from network theory, *J. Non-Newtonian Fluid Mech.* **2**, 353.
- Phan-Thien, N., 1978, A nonlinear network viscoelastic model, *J. Rheol.* **22**, 259.
- Silagy, D., Y. Demay, and J.-F. Agassant, 1996, Study of the stability of the film casting process, *Polym. Eng. Sci.* **36**, 2614.
- Yoon, K.-S., and C.-W. Park, 2000, Stability of a two-layer blown film coextrusion, *J. Non-Newtonian Fluid Mech.* **89**, 97.