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Stability analysis of a three-layer film casting process

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Abstract

The co-extrusion of multi-layer films has been studied with the focus on its process stability. As in the single-layer film casting process, the productivity of the industrially important multi-layer film casting and the quality of thus produced films have often been hampered by various instabilities occurring in the process including draw resonance, a supercritical Hopf bifurcation instability, frequently encountered when the draw ratio is raised beyond a certain critical value. In this study, this draw resonance instability along with the neck-in of the film width has been investigated for a three-layer film casting using a varying width non-isothermal 1-D model of the system with Phan-Thien and Tanner (PTT) constitutive equation known for its robustness in portraying extensional deformation processes. The effects of various process conditions, e.g., the aspect ratio, the thickness ratio of the individual film layers, and cooling of the process, on the stability have been examined through the nonlinear stability analysis.

Keywords : draw resonance, film casting, multi-layer film, stability, transient solutions

1. Introduction

The co-extrusion process involves bringing together two or more polymeric streams, often with quite different rheological properties and melt temperatures, and extruding a multi-layered structure through a co-extrusion die (Fig. 1). Despite its design and operation complexities, the co-extrusion process making multi-layer films or sheets has steadily been advanced as an important plastic fabrication process (Pis-Lopez and Co, 1996; Khomami and Su, 2000; Valette *et al.*, 2004), to satisfy the customers' various needs such as inserting barriers, placing colors, burying recycled materials, coatings for controlling film surface properties, etc. Most of coextruded films can be manufactured through either the film casting or the tubular film blowing: among them the film casting being preferred for precise processing operations because of its refined control of process conditions like die extrusion and efficient film cooling on chill roll and of better resulting film qualities like optical clarity (Kanai and Campbell, 1999).

In the multi-layer film casting process, multiple melt streams from different extruders are combined in a feed block, which produces a multi-layer melt. During the cast film co-extrusion process, the resin is extruded through a flat slit onto a chill roll. The rotation speed of that roll controls the draw ratio and the final thickness of the film. Several types of instabilities and defects, however, limit the productivity of this process: interfacial instabilities of either high frequency, zig-zag or long wave type occurring in the pre-die region (Khomami and Su, 2000; Valette *et al.*, 2004), and film neck-in, edge beads, and draw resonance arising in the post-die region (Silagy *et al.*, 1998; Kim *et al.*, 2005; Shin *et al.*, 2007). In this study, the draw resonance that has historically attracted many researchers' interest over the last four decades as an industrially important productivity issue as well as an academically interesting stability topic and is characterized by the periodic oscillations of film thickness and width has been extensively investigated (Pearson and Matovich, 1969; Fisher and Denn, 1976; Hyun, 1978; Lee *et al.*, 2001; Jung *et al.*, 2004; Jung and Hyun, 2005, 2006). A three-layer film casting process, i.e., A-B-A type, has been selected as an example to analyze this draw resonance instability along with transient responses of the non-isothermal system toward various other process conditions, using a varying width 1-D model with a realistic constitutive equation capable of portraying the behavior of both extensional thickening and thinning fluids.

2. Modeling

The varying width 1-D model by Silagy *et al.* (1996) has been coupled with the constant width 1-D multi-layer model by Pis-Lopez and Co (1996) and an energy equation to investigate the dynamics and stability of non-isothermal

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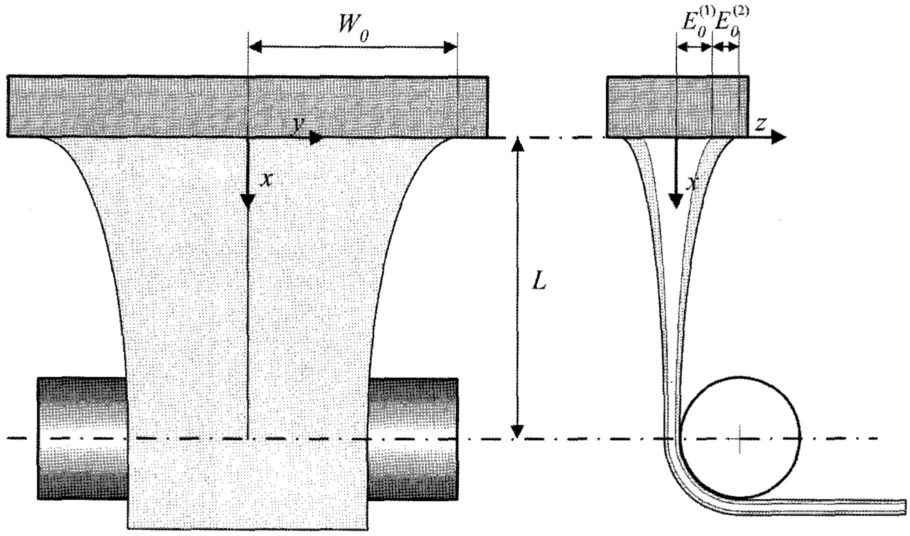


Fig. 1. Schematic diagram of a three-layer film casting process.

multi-layer film casting including the neck-in phenomenon which couldn't be explored by the earlier constant width 1-D model (Anturkar and Co, 1988; Iyengar and Co, 1996). Under the assumptions that velocity gradients are constant along the film width direction, reduction of film width (neck-in) can be approximately described even in 1-D model (Silagy *et al.*, 1996, Lee *et al.*, 2001). It has been proved that this assumption is so effective to qualitatively portray neck-in phenomena (Lee *et al.*, 2004a). Particularly in the present study, a three-layer (A-B-A type) film casting is investigated for both cases where A is a bulk component with B a minor, and where B is a bulk with A minor. The former represents the cases when a barrier layer, B, is inserted into the main film, A, whereas the latter is when a film, B, is coated with film layers, A, at both sides. The Phan-Thien and Tanner (PTT) constitutive fluid model was employed (Phan-Thien, 1978), and values of many parameters in the system were adopted from the literature. The configuration of the system is delineated in Fig. 1 and the dimensionless governing equations are as follows.

Equation of continuity:

$$\frac{\partial[e^{(1)}w]}{\partial t}+\frac{\partial[e^{(1)}wv]}{\partial x}=0,\ \frac{\partial[e^{(2)}w]}{\partial t}+\frac{\partial[e^{(2)}wv]}{\partial x}=0 \qquad (1)$$

where, $e^{(k)}\equiv\frac{E^{(k)}}{E_0},\ w\equiv\frac{W}{W_0},\ v\equiv\frac{V}{V_0},\ t\equiv\frac{\bar{t}V_0}{L},\ x\equiv\frac{X}{L}$

Equation of motion: $F=\varsigma^{(1)}\sigma_{xx}^{(1)}e^{(1)}w+\varsigma^{(2)}\sigma_{xx}^{(2)}e^{(2)}w \qquad (2)$

where, $\sigma_{ij}^{(k)}\equiv\frac{\bar{\sigma}_{ij}^{(k)}L}{2\eta_0^{(1)}V_0},\ \varsigma^{(k)}\equiv\frac{\eta^{(k)}}{\eta^{(1)}}$

Constitutive equation for k^{th} layer (PTT model):

$$K^{(1)}\boldsymbol{\tau}^{(1)}+\mathrm{De}^{(1)}\left[\frac{\partial\boldsymbol{\tau}^{(1)}}{\partial t}+v\cdot\nabla\boldsymbol{\tau}^{(1)}-\boldsymbol{L}^{(1)}\cdot\boldsymbol{\tau}^{(1)}-\boldsymbol{\tau}^{(1)}\cdot\boldsymbol{L}^{(1)^T}\right]=\frac{\mathrm{De}^{(1)}}{\mathrm{De}_0^{(1)}}\boldsymbol{D},$$

$$K^{(2)}\boldsymbol{\tau}^{(2)}+\mathrm{De}^{(2)}\left[\frac{\partial\boldsymbol{\tau}^{(2)}}{\partial t}+v\cdot\nabla\boldsymbol{\tau}^{(2)}-\boldsymbol{L}^{(2)}\cdot\boldsymbol{\tau}^{(2)}-\boldsymbol{\tau}^{(2)}\cdot\boldsymbol{L}^{(2)^T}\right]=\frac{\mathrm{De}^{(2)}}{\mathrm{De}_0^{(2)}}\boldsymbol{D} \qquad (3)$$

where, $\mathrm{De}_0^{(k)}\equiv\frac{\lambda_0^{(k)}V_0}{L},\ \mathrm{De}^{(k)}\equiv\mathrm{De}_0^{(k)}\exp\left[\frac{E_a^{(k)}}{RT_0}\left(\frac{1}{\theta^{(k)}}-1\right)\right],\ \tau_{ij}^{(k)}\equiv\frac{\bar{\tau}_{ij}^{(k)}L}{2\eta^{(1)}V_0},$

$K^{(k)}=\exp[2\varepsilon^{(k)}\mathrm{De}_0^{(k)}\mathrm{trace}[\boldsymbol{\tau}^{(k)}]],$

$\boldsymbol{L}^{(k)}=\nabla v-\xi^{(k)}\boldsymbol{D},\ 2\boldsymbol{D}=[(\nabla v)+(\nabla v)^T]$

Equation of energy: $\frac{\partial\theta^{(1)}}{\partial t}+v\frac{\partial\theta^{(1)}}{\partial x}=\mathrm{St}^{(1)}\frac{[\theta^{(2)}-\theta^{(1)}]}{e^{(1)}}$

$$\frac{\partial\theta^{(2)}}{\partial t}+v\frac{\partial\theta^{(2)}}{\partial x}=\mathrm{St}^{(2)}\frac{[\theta_a-\theta^{(2)}]}{e^{(2)}} \qquad (4)$$

where, $\theta^{(k)}\equiv\frac{T^{(k)}}{T_0^{(1)}},\ \mathrm{St}^{(k)}\equiv\frac{2h^{(k)}L}{\rho^{(k)}C_p^{(k)}V_0E_0}$

Edge condition: $[q\sigma_{xx}^{(1)}+(1-q)\sigma_{xx}^{(2)}]\left(\frac{\partial w}{\partial x}\right)^2=A_r^{\,2}[q\sigma_{yy}^{(1)}+(1-q)\sigma_{yy}^{(2)}] \qquad (5)$

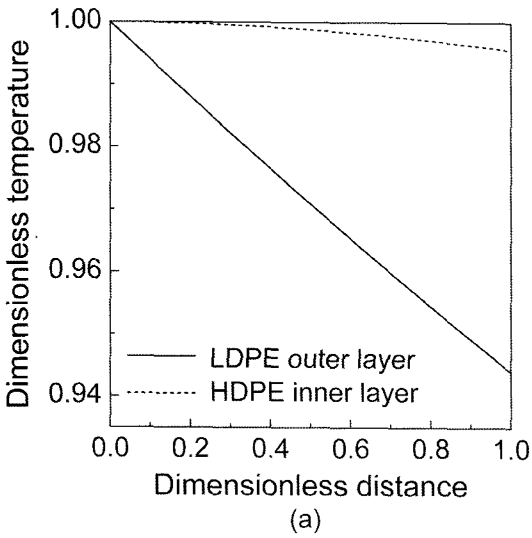
where, $A_r\equiv\frac{L}{W_0}$

Boundary conditions:

$t=0$: $e_0^{(1)}=q,\ e_0^{(2)}=1-q,\ w_0=1,\ \theta_0^{(k)}=1,\ v_0=1,\ v_L=D_r \qquad (6a)$

$t>0$: $e_0^{(1)}=q,\ e_0^{(2)}=1-q,\ w_0=1,\ \theta_0^{(k)}=1,\ v_0=1,\ v_L=D_r(1+\delta) \qquad (6b)$

where e, w, v, t, θ and x are the dimensionless variables of film thickness, film width, velocity in flow direction, time, temperature, and distance in flow direction, respectively. F, τ_{ij}, and σ_{ij} denotes the dimensionless axial tension, extra stress component in ij-direction, and total stress tensor in ij-direction, respectively. De the Deborah number, ε, ξ the PTT model parameters, E_a the activation energy, R the gas constant, T_0 the extrusion temperature, St the Stanton num-



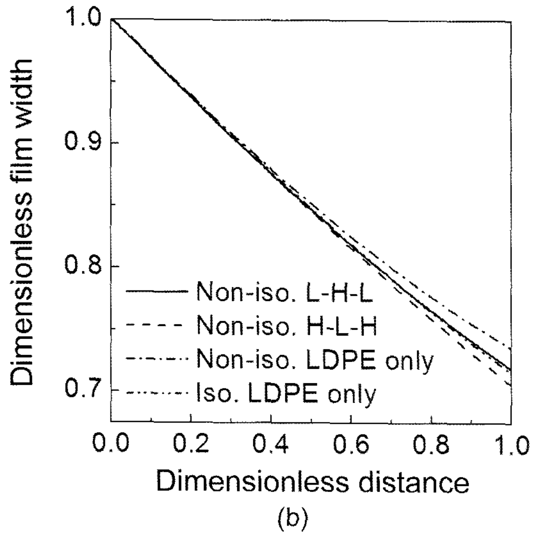


Fig. 2. (a) Dimensionless temperature profiles for the L-H-L film casting at $A_r=0.5$, $D_r=20$ and $\varsigma=1.0$ and (b) dimensionless film width profiles of single-layer and three-layer film castings.

ber, h the heat transfer coefficient, ρ the fluid density, C_p the heat capacity, θ_a the dimensionless ambient temperature, q inner layer film thickness at die exit, and δ the constant initial step disturbance at the take-up velocity. Superscript k represents the k^{th} layer from center of the film and subscripts 0 and L denote die exit and take-up positions, respectively. Five different ratios, i.e., the thickness ratio ($\varphi\equiv e_0^{(2)}/e_0^{(1)}$), the viscosity ratio ($\varsigma\equiv\eta^{(2)}/\eta^{(1)}$), the elasticity ratio ($\Lambda\equiv\lambda^{(2)}/\lambda^{(1)}\equiv De^{(2)}/De^{(1)}$), the aspect ratio ($A_r\equiv L/W_0$), and the drawdown ratio ($D_r\equiv V_L/V_0$), are the essential elements of the governing equations of the A-B-A layer film casting process in this study. For the edge condition in Eq. (5), it is assumed the stresses of each layer equally contributed on the edge free surface. Due to the limit of 1-D model adopted in this study, each layer has the same film width (w) and flow velocity (v). In addition, film temperature is simply averaged in each layer because the temperature distribution could not be evaluated in film thickness direction. (Temperature in the outer layer is lower than in the inner layer due to the cooling by ambient air (Fig. 2a).)

Finite difference method (FDM) with 1,000×5,000 meshes in x-t grid employed in this study guarantees the numerical accuracy in transient solutions, after a 5% step change in take-up velocity as an initial disturbance to the system, as well as steady states. The spatial coordinate was discretized by the backward difference scheme with the 2nd-order accuracy, and absolutely stable 2nd-order Gear method (so-called BDF2) was incorporated for time discretization to minimize any possible numerical instabilities. It turns out that this numerical scheme here is superior to 1st-order backward method with 2,000×10,000 meshes in x-t grid (Deville *et al.*, 2002). (In order to check the results in multi-layer film casting, neck-in profiles obtained by varying width 1-D isothermal (Lee *et al.*, 2001) and non-isothermal models have been compared in Fig. 2b, demonstrating physically affordable behavior according to the viscoelasticity of the materials and cooling conditions.)

3. Results and discussion

The capability of the simulation model of Eqs. (1)-(6) in determining the stability of the three-layer film casting process has been tested in this study, changing the parameters and processing conditions, for example LDPE and HDPE systems. To demonstrate the dichotomous behavior of poly-

Table 1. Model parameters used in this study

Parameters	LDPE	HDPE
Melt density, ρ (kg/m^3) at 473K[a]	755	755
Heat capacity, C_p (J/kg K)[b]	2260	2260
Heat transfer coefficient, h (J/m^2·K·s)[c]	7.0	7.0
Relaxation time, λ_0 (s)[a]	0.05	0.02
PTT model parameters, ε & ξ[a]	0.015, 0.1	0.015, 0.7
Activation energy, E_a (kJ/mol)[d,e]	52.21	27.24
Extrusion velocity, V_0 (mm/s)[f]	5.0	
Half width of T-die (mm)[f]	50	
Distance from die to chill roll (mm)[f]	25	
Extrusion temperature, T_0 (K)[f]	473.15	

Sources
- a. Typical material data
- b. Van Krevelen, D.W., 1990, Properties of Polymers.
- c. Smith, S. and D. Stolle, 2000, *Polym. Eng. Sci.* **40**, 1870.
- d. Satoh, N., 2001, *Polym. Eng. Sci.* **41**, 1564.
- e. Dealy, J.M. and K.F. Wissbrun, 1990, Melt Rheology and Its Role in Plastics Processing.
- f. General operating conditions of our table-top scale film casting apparatus.

mer melts according to their extensional characteristics, the PTT model parameters are set to $\varepsilon^{(k)}$=0.015, $\xi^{(k)}$=0.1 for LDPE and $\varepsilon^{(k)}$=0.015, $\xi^{(k)}$=0.7 for HDPE, respectively (Phan-Thien, 1978; Lee *et al.*, 1995; Shin *et al.*, 2007). Typical data for other rheological properties and model parameters used here are summarized in Table 1 (Dealy and Wissbrun, 1990; Kanai and Campbell, 1999; Smith and Stolle, 2000; Satoh, 2001). Long chain branched polymers typically possess a higher activation energy (E_a) for the temperature dependency of viscosity than linear polymers with the same chemical structure. E_a is changed from about 27.3 kJ/mol·K for the linear polymers up to about 58.8 kJ/mol·K for branched polymers (Dealy and Wissbrun, 1990). It has been manifested that the larger quantity of LDPE as compared with that of HDPE in the three-layer system, the more stable the system becomes, irrespective of L-H-L or H-L-H system. In other words, increasing portion of the extension thickening material in multi-layer film makes the system more stable. Fig. 3 displays the simulation results of the stability enhancement of both L-H-L and H-L-H systems by increasing LDPE portion at the die exit. It can be shown in Fig. 3 that the process stability is quite different, according to the placement of LDPE (or HDPE) in outer or inner region.

The above findings are not surprising in view of the fact that extension thickening or strain hardening material, LDPE, exhibits more stable behavior than extension thinning or strain softening one, HDPE, in extension deformation processes such as fiber spinning and single-layer film casting (Pis-Lopez and Co, 1996; Lee *et al.*, 2003). LDPE generally provides excellent extrusion processability and ensures the high draw ratio condition, whereas LLDPE and HDPE are known to have relatively poor extrusion processability. From the stability results of single-layer film casting process, it has also been confirmed that the extension thickening fluid, e.g., LDPE with long chain branched structure is more stable than extension thinning fluid like linear structured HDPE (White and Ide, 1978;

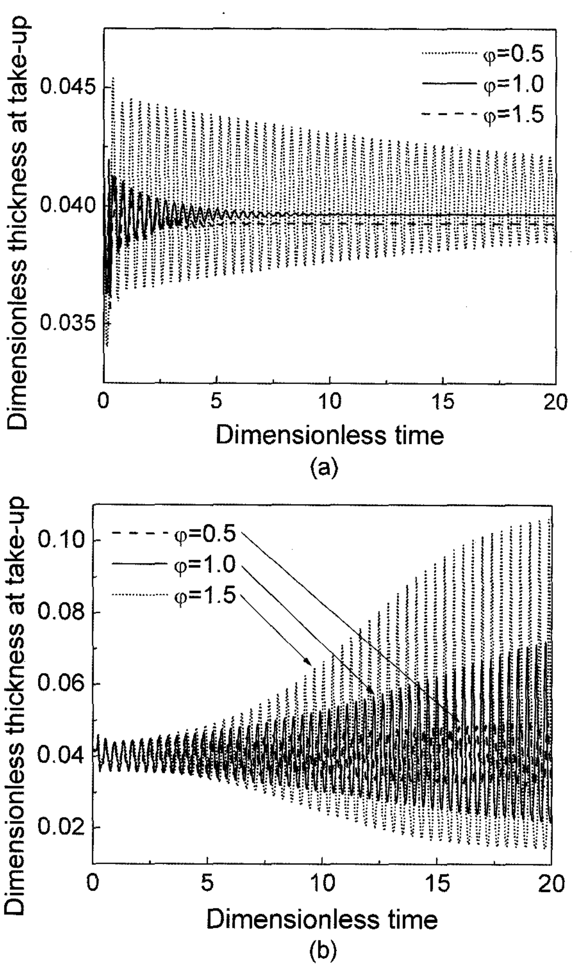


Fig. 3. Effect of thickness ratio, φ, on the process stability using transient responses for (a) L-H-L and (b) H-L-H systems at A_r=0.5, D_r=35 and ς=1.0.

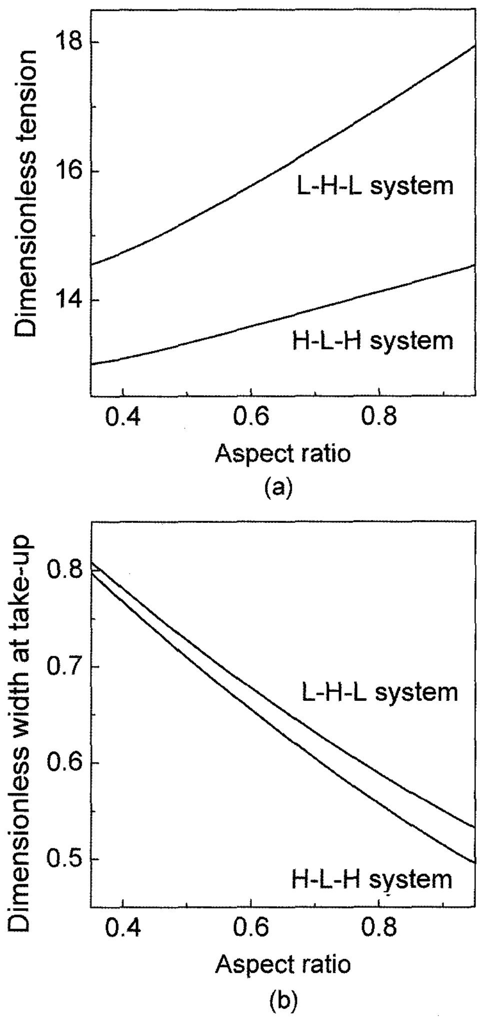


Fig. 4. Effect of aspect ratio on (a) the axial tension and (b) the reduction of film width (neck-in) in L-H-L and H-L-H systems at A_r=0.5, D_r=30 and ς=1.0.

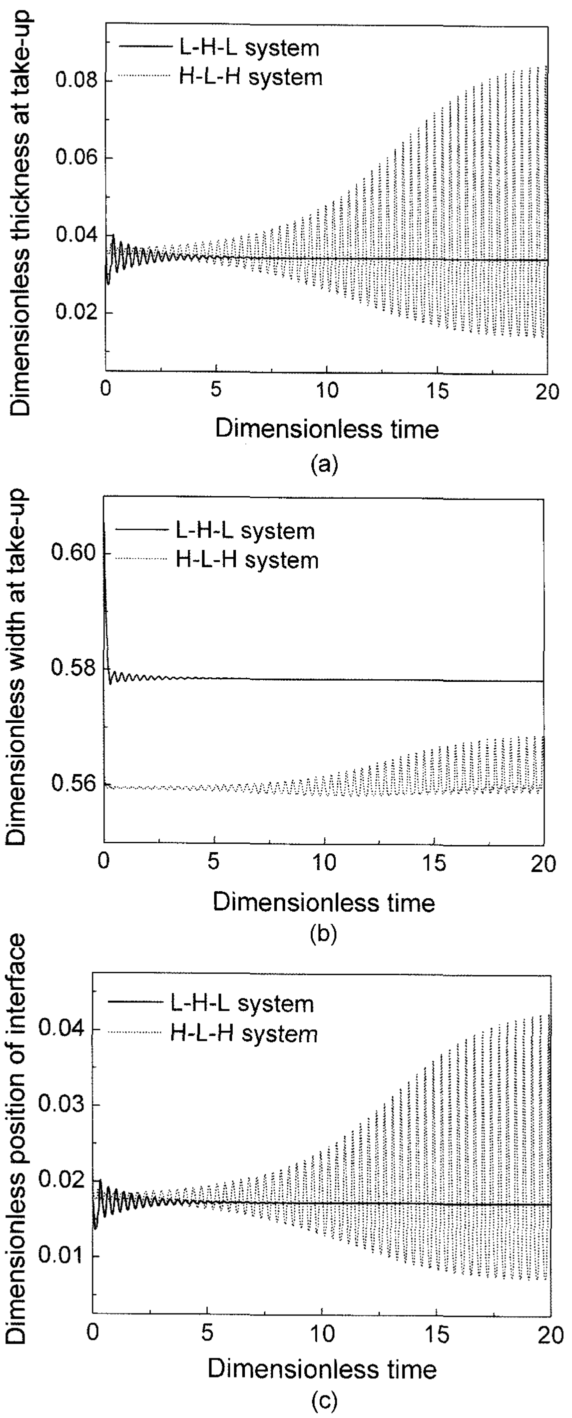


Fig. 5. Transient responses of (a) the film thickness, (b) the film width, and (c) the interface position in L-H-L and H-L-H systems ($\varphi = 1.0$, A_r=0.8, D_r=50 and $\varsigma = 1.0$).

Shin *et al.*, 2007). Moreover, fluid viscoelasticity (De) stabilizes the process for the extension thickening fluid case, whereas the extension thinning fluid case is the opposite (Lee *et al.*, 2003). Such a dichotomous behavior on the process stability, depending on the extensional properties of polymer melts, is clearly observed in other extensional deformation processes as mentioned above, such as fiber spinning and film blowing (Lee *et al.*, 1995; Ghaneh-Fard *et al.*, 1996).

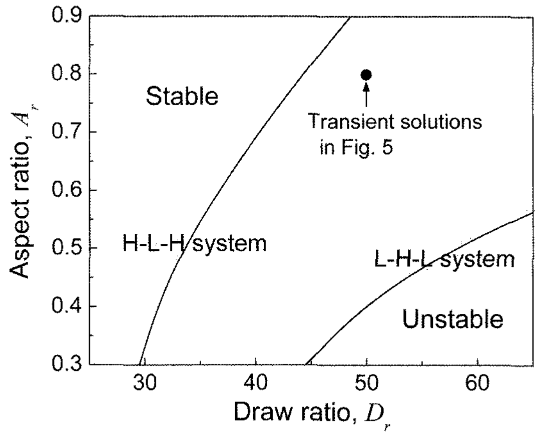


Fig. 6. Stability diagram of two systems in Fig. 5 on draw ratio, D_r and aspect ratio, A_r, plane.

Fig. 4 shows that the axial tension and neck-in increase with aspect ratio in both L-H-L and H-L-H systems. The reason that axial tension and film width of L-H-L case are higher than in the H-L-H case comes from the difference of cooling condition acting onto the film. As will be explained later, LDPE in L-H-L system is strongly affected by cooling temperature, leading to high axial tension and reduced neck-in due to the enhanced viscoelasticity (Shin *et al.*, 2007).

Other interesting results can be clarified when outer and inner layers of the multi-layer system are shifted each other. Fig. 5 depicts transient simulation results for both cases of L-H-L and H-L-H. The position of interface can be also captured along with the evolution of time, showing the same oscillation period and dynamic behavior as those of the total film thickness and film width. To ascertain this stability change of the three-layer film casting by the selection of outer material, the neutral stability curves are plotted on the D_r-A_r plane in Fig. 6. For the isothermal models, the optimal aspect ratio, ensuring maximum stable region, is found around A_r=1.5 for the varying width 1-D model (Silagy *et al.*, 1996; Lee *et al.*, 2003) and A_r=0.8 for 2-D model (Kim *et al.*, 2005). However, it is expected that this optimal aspect ratio does not appear in the non-isothermal model due to the cooling between die to chill roll up to higher aspect ratio regime. As the aspect ratio increases, the process is more stabilized because of the strong cooling acting in the extended flow distance (L). Difference of onsets in both cases becomes wider as increasing A_r, as shown in Fig. 6. It has been noted that it is important to select the outer material for the stabilization of the multi-layer system by effective cooling, i.e., the system with higher E_a guarantees the better processability, especially for the extensional thickening materials. As already known to the previous studies (Lucchesi *et al.*, 1985; Jung *et al.*, 1999), the cooling condition can effectively stabilize the process and then, it depends on properties of an outer film directly facing on ambient air. If the long chain branched LDPE film with higher activation energy than HDPE (Table 1) is employed as outer layer under the same operating conditions, the tension applied on molten film more drastically increased due to cooling, making the L-H-L system more stable.

Consequently, to operate the three-layer film casting in the stable region, two strategies are worth mentioning, i.e., to enlarge the portion of the extension thickening material like LDPE and to choose temperature sensitive material as the outer film. Also, other process conditions such as viscosity ratio (ς) and elasticity ratio (Λ) in this three-layer system have been considered, providing similar stability tendency (Lee *et al.*, 2004b). Sensitivity results of the multi-layer system to any disturbances can be explained using tension sensitivity analysis developed in Jung *et al.* (1999), e.g., cooling makes the multi-layer system less sensitive to any disturbances. From the above stability results, the origin of ingenious devices such as draw resonance eliminator (Lucchesi *et al.*, 1985) and encapsulation dies (Lee *et al.*, 2004c) can be easily understood and thus new equipment can be further developed to control instabilities, inevitably occurring in extensional deformation processes.

4. Conclusions

Nonlinear stability of the multi-layer film casting process has been analyzed using a non-isothermal varying width 1-D model employing a Phan-Thien and Tanner constitutive equation. It is well known that the long chain branched polymers can enhance the processability of polymer processing by virtue of their extensional hardening characteristics. In the case of draw resonance, the portion of extension thickening material and its fluid viscoelasticity in multi-layer system alleviate the instability and ultimately stabilize the system, whereas those of the extension thinning material aggravate the instability. In addition to the extensional properties of the materials, the higher activation energy of long chain branched polymers, albeit the same chemical structures, contributes to stabilize the film casting process. Especially for the multi-layer system, temperature sensitive material should be selected as the outer film to improve the process stability by imposing maximum tension of the film. These stability results can be also corroborated through the sensitivity analysis that has been successfully applied to the single-layer film casting process.

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