Article

Relationship between Neck-in Phenomena and Rheological Properties in Film Casting

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We numerically demonstrate that the ratio of uniaxial to planar elongational viscosity controls the neck-in phenomena of the casting polymer films, i.e., the decrease in film width value for both viscous and viscoelastic fluids. Quasi-three-dimensional numerical simulations of an isothermal film casting process were performed using a finite element method for two viscoelastic fluids using Larson and PTT models, purely-viscous non-Newtonian fluids using Cross model and Newtonian fluids. The increase in take-up velocity relative to that in extrusion showed particular film deformations of neck-in depending upon the rheological properties in the models. The results indicate that the film width is determined by the ratio of uniaxial to planar elongational viscosity rather than the extension-thickening nature in uniaxial elongation. The computations using different fluid models show that the viscosity ratio is universal for predicting the neck-in value of the stretching polymer films not only in viscoelastic but also in pure viscous fluids.

Key Words: Film casting / Neck-in / Elongational viscosity / Viscoelastic fluid / Viscous fluid

1. INTRODUCTION

In industrial film casting, the molten polymer is extruded through a flat die and taken up by a rotating chill/nip roll to form a thin polymer film as schematically shown in Fig. 1. The greater take-up velocity at the chill roll than that in extrusion causes the film to be deformed not only in the longitudinal but also in transverse and thickness directions, resulting in the two characteristic deformation defects, i.e., the “neck-in” and “edge-bead”. The former represents the decrease in film width along the flow direction due to a preferential deformation in the transverse direction, while the latter is defined as a film thickening at the edge. From a practical point of view, it is desirable to suppress these deformations in order to achieve a uniform film thickness with high productivity.

Successful physical models have been presented for describing the physical mechanism of the film deformation in the casting process. The simplest case is for Newtonian fluid involving neither elasticity nor rate-dependent properties. d’Halewyn et al.1) proposed a quasi-three-dimensional numerical model for an isothermal Newtonian fluid. The velocity and stress gradient in the thickness direction were assumed to be negligible compared to those in the other directions. It has been demonstrated that this simplified model successfully showed a particular neck-in and edge-bead

Fig. 1. Schematic of the film casting process from a flat die.

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phenomena. A complete three-dimensional flow simulation of the film casting process was first performed by Sakaki et al.\(^2, 3\) for Newtonian fluid under different take-up velocity and air gap length conditions. The simulated film width was in good agreement with the experimental data for a linear high-density polyethylene (HDPE).\(^3\)

For viscoelastic fluids, the neck-in value, defined as the difference between the die width and the film width, depends upon not only the operating conditions but also upon material properties of polymers such as elongational viscosities, relaxation times and relaxation modulus. Debbaut et al.\(^4\) showed the quasi-three-dimensional simulations using Upper Convective Maxwell model and Giesekus model with a single relaxation time. They suggested that the calculated film width increased with increasing uniaxial elongational viscosity, i.e., with increasing extension-thickening nature in uniaxial elongation. The same extension-thickening dependence of the film width has been demonstrated by Satoh et al.\(^5\) from the quasi-three-dimensional simulations using Larson models with multiple relaxation times. They also compared the numerical computations with laboratory experiments, indicating that the computed film width agreed with the measured data for four kinds of branched low-density polyethylene (LDPE) exhibiting different uniaxial elongational viscosities.

In contrast to the extensive numerical studies on the film casting for Newtonian and non-Newtonian fluids, the key material property that dominates the neck-in phenomenon has not yet been clarified. Most previous studies have implied that the extension-thickening nature in uniaxial elongation is an important physical factor for the transverse film deformation. A branched LDPE showed a smaller neck-in value due to its stronger extension-thickening property,\(^5\) while a linear HDPE exhibiting smaller uniaxial elongational viscosity leads to a larger neck-in.\(^3\) However, the extension-thickening property in uniaxial elongation would dominate the film deformation only in the case when the film elongates in uniaxial stress at all transverse positions. Debroth and Erwin\(^6\) and Satoh et al.\(^5\) have suggested that the film edge elongates in uniaxial stress while the film center does in plane stress. Because the planar elongational viscosity rather than the uniaxial one is expected to play an important role for the film deformation at the film center, it is important to take into account both the uniaxial/planar elongation kinetics in order to understand the essential mechanism of the polymer deformations in the film casting. Ito et al. have also performed the experimental flow analysis and have derived the analytic equation for the prediction of neck-in value.\(^7, 8\) They point out that neck-in length is determined by the ratio of planar elongational viscosities, \(\eta_{pl}/\eta_{p2}\), where \(\eta_{pl}\) is derived from normal stress difference between stretching and squeezing directions while \(\eta_{p2}\) between width and squeezing directions.

In this paper, we proposed a new rheological property, i.e., the ratio of uniaxial to planar elongational viscosity, in order to estimate the neck-in value in film casting process for viscous and viscoelastic fluids. If the neck-in value is determined by this index, the results should be valid whether the material is viscous or viscoelastic. The numerical investigation using the flow analysis is suitable for the purpose because the experimental verification is difficult. We performed the quasi-three-dimensional isothermal simulations with reliable assumptions, which were confirmed in the previous studies as described in next chapter, for two viscoelastic fluids using Larson and Phan-Thien/Tanner (PTT) models, purely-viscous fluids using Cross model and Newtonian fluids. Single relaxation models were employed for the viscoelastic fluids to clarify the detailed physical mechanism of the neck-in phenomenon. The results indicated that film width was determined by the viscosity ratio for both the viscoelastic and viscous models. The film width decreased with decreasing viscosity ratio even for high uniaxial elongational viscosities, suggesting that the extension-thickening nature in uniaxial elongation is not the universal factor for the film neck-in.

### 2. GOVERNING EQUATIONS AND BOUNDARY CONDITIONS

The quasi-three-dimensional flow analysis used in the present study is based on D’Hallewyn et al.\(^1\) The velocity and stress gradient in the thickness direction are assumed to be negligible compared to those in the other directions. The validity of this assumption has been verified by Satoh et al.\(^5\) by comparing the numerical results of fully-three-dimensional simulations. The effect of gravity force can be negligible over 2 in draw ratio.\(^2, 3\) The continuity and momentum equation are integrated across the film thickness and given by:

\[
\frac{\partial}{\partial x} (e v_x) + \frac{\partial}{\partial y} (e v_y) = 0
\]

(1)

\[
\frac{\partial}{\partial x} (\sigma_{xx}) + \frac{\partial}{\partial y} (\sigma_{xy}) = 0
\]

(2)

\[
\frac{\partial}{\partial x} (\sigma_{yy}) + \frac{\partial}{\partial y} (\sigma_{yy}) = 0
\]

(3)
where \( e = e(x,y) \) is the film thickness and \( v_i \) \((i = x \text{ or } y)\) is the \( i \)-component of velocity vector \( v \). \( \sigma_{ij} \) \((i,j = x \text{ or } y)\) is the \( ij \)-component of total stress tensor \( \sigma \) defined as

\[
\sigma = -p\delta + \tau
\]

where \( p \) is the hydrostatic pressure, \( \tau \) extra-stress tensor and \( \delta \) the unit tensor. The three-dimensional shape of the film was obtained by solving these equations simultaneously.

The following four constitutive equations were used for the computation. The Larson and PTT models involve viscoelastic natures while Newtonian and Cross models involve no elasticity.

**Newtonian:**

\[
\tau = 2\eta D
\]

**Cross:**

\[
\eta\Pi = \frac{\eta_0}{1 + C(\lambda_0\Pi)^{\gamma_n}}
\]

**Larson:**

\[
\tau + \lambda D \tau + \frac{2}{3}G D : \sigma = 2\eta D
\]

**PTT:**

\[
\left[ \exp \left( \frac{\zeta}{G} \text{tr}(\tau) \right) \right] \tau + \frac{\zeta}{2} \text{tr}(\tau) \left( 1 - \frac{\zeta}{2} \right) \tau = 2\eta D
\]

where \( \lambda \) is the relaxation time, \( G \) the relaxation modulus, \( D \) the rate of strain tensor, \( \Pi \) the second invariant of \( D \), and \( \zeta \), \( \epsilon \) and \( \xi \) are the nonlinear parameters contained in Larson and PTT models. \( C \) and \( \lambda_0 \) are the parameters in the Cross model. \( \gamma \) and \( \Delta \) denote the upper and lower convected time derivatives, respectively. Although most polymer melts generally involve multiple relaxation times, single relaxation models were employed for these viscoelastic fluids in order to clarify the detailed physical mechanism of the neck-in phenomenon.

The boundary conditions at the die exit and the chill roll are given by:

**Die exit:**

\[
v_i = v_1, \quad v_j = 0, \quad \tau = \tau_1
\]

**Chill roll:**

\[
v_i = v_2, \quad v_j = 0
\]

where the values \( v_1 \) and \( v_2 \) are the extrusion velocity and the take-up velocity. Draw ratio is defined as the ratio of \( v_2 \) to \( v_1 \). The stress at the die exit, \( \tau_1 \), was determined assuming an analytical solution for Newtonian fluid.\(^5\) It was confirmed that the final film shape at the chill roll was not influenced by the boundary conditions at the die exit.\(^2\,\,^3\) The die width, die thickness and air gap used in the computations were 250 mm, 0.8 mm and 88 mm, respectively.

At the free surface, the surface tension and air drag were assumed to be negligible compared to the internal viscous and elastic stresses.

\[
n \cdot v = 0
\]

\[
n \cdot \sigma = 0
\]

where \( n \) is the normal unit vector perpendicular to the free surface.

The Galerkin finite element method (GFEM) was used to solve the velocity and film thickness fields. The velocity and film thickness in each GFEM mesh were approximated by means of biquadratic interpolation functions. The stress field was solved by integrating the constitutive equation along the streamlines using Euler integration method. For the viscoelastic case, the Newtonian solutions were used as the initial guess of the exact solutions. A detailed description of the numerical procedure is available elsewhere.\(^5\)

### 3. RESULTS AND DISCUSSION

#### 3.1 Larson Model

Figure 2 shows the elongational viscosities as a function of elongation rate for three different viscoelastic Larson models. The solid and broken curves represent the viscosities of

![Fig. 2. Variation in uniaxial and planar elongational viscosities with elongation rate for Larson model. The solid and broken curves represent the viscosities of uniaxial and planar elongation, respectively. The viscosities are respectively normalized by zero-elongation-rate viscosity of \( 3\eta_0 \) for the uniaxial viscosity, and \( 4\eta_0 \) for the planar viscosity. Relaxation times and nonlinear parameters in the constitutive equation are chosen to involve strong (Model A fluid) and weak (Models B and C) extension-thickening properties.](image-url)
uniaxial and planar elongation, respectively. The viscosities are respectively normalized by zero-elongation-rate viscosity of $3\eta_0$ for the uniaxial viscosity, and $4\eta_0$ for the planar viscosity. Model A fluid shows typical extension-thickening properties due to a longer relaxation time and a smaller nonlinear parameter, i.e., stronger interactions between polymer chains. In contrast, Models B and C indicate weaker extension-thickening natures due to the larger value of the nonlinear parameter. The longer relaxation time in Model C fluid leads to a decrease in elongation rate of nonlinear region. The physical parameters involved in these models were chosen to show the above-mentioned rheological characteristics in elongation.

If the extension-thickening property is the major factor that dominates the neck-in phenomenon, Model A fluid is expected to have the largest film width compared to other two models. The calculated film width normalized by the die width is shown in Fig. 3 as a function of draw ratio, defined as the take-up velocity to the inlet velocity of the fluid. The draw ratio ranging from 1 to 40 corresponds to the elongation rates of $0 - 3\, \text{s}^{-1}$. It is found from Fig. 3 that the Model C fluid exhibiting the weaker extension-thickening nature shows the larger film width, i.e., smaller neck-in value, than the Model A fluid. This fact provides conclusive evidence that the neck-in value is not determined by the extension-thickening property of viscoelastic fluids.

Figure 3 also reveals that the calculated film width first decreases and then slightly increases with increasing draw ratio. The increase in film width in the high draw ratio regime is a characteristic nature of fluids exhibiting the extension-thickening property. The film width in Model A fluid was found to be only 4% larger than that in Model B at the draw ratio of 40, which is much smaller than the difference in film width between Models C and A. Because the uniaxial elongational viscosity in Model A is more than 40% larger than that in Model B at the largest draw ratio, this provides another evidence that the film width involves few correlation with the extension-thickening nature of fluids.

Here we assume that the ratio of normalized uniaxial elongational viscosity $\eta_{EU}/3\eta_0$ to the planar elongational viscosity $\eta_{EU}/4\eta_0$ is the essential material property for the neck-in defect formation. As mentioned in Introduction, the film edge elongates in uniaxial stress while the film center does in plane stress. Thus it is reasonable to expect that the neck-in phenomenon originates from a subtle balance of the uniaxial/planar elongational deformations, that is, the relative importance of uniaxial and planar viscosities at a given elongation rate. Figure 4 shows the variation in viscosity ratio with the elongation rate for the three Larson models. The viscosity ratio is unity in low elongation rate regimes, corresponding to a particular Newtonian behavior. It is found that the Model C exhibiting the largest film width (see Fig. 3) indicates the highest viscosity ratio for a wide range of elongation rates from $10^{-3}$ to $5\, \text{s}^{-1}$. Models A and B show almost the same viscosity ratio as does the film width in Fig. 3. These facts suggest that the neck-in value is determined by the ratio of uniaxial to the planar elongational viscosity under the same operating conditions.

![Fig. 3. Variation in film width with draw ratio for Larson Model. The solid, dotted and broken curves represent the film width predicted from Models A, B and C respectively. The draw ratio is defined as the take-up velocity to the inlet velocity of the fluids.](image1)

![Fig. 4. Variation in ratio of uniaxial to planar elongational viscosity with elongation rate for Larson Model. The solid, dotted and broken curves represent the film width predicted from Models A, B and C respectively.](image2)
This particular dependence of film width upon the viscosity ratio can be understood in terms of the competing elongational deformations at the film edge and the center. Suppose that the planar elongational viscosity of the polymer is low compared to that in uniaxial elongation. The resulting planar elongational deformation at the film center would suppress the film shrinkage in the transverse direction due to a preferential deformation in the thickness direction. At the same time, the relatively higher uniaxial elongational viscosity can retard deformations both in the transverse and thickness directions at the film edge. These two specific contributions for the film deformations lead to a larger film width at the greater viscosity ratio as shown in Fig. 3 (Model C). On the contrary, the lower viscosity ratio of the fluid favors the transverse film deformation along the flow direction, resulting in a smaller film width.

3.2 PTT Model

We showed in the previous section that the ratio of uniaxial to planar elongational viscosity was a key material property for a film casting of viscoelastic Larson fluid. Because any fluids exhibit their characteristic viscosity ratios, it would be reasonable to expect that other viscous/viscoelastic models would show a specific neck-in value depending upon the viscosity ratios of the fluids. In the following sections, we show the universality of the viscosity ratio to other fluids using the viscoelastic PTT model and viscous Cross model.

First, we will assure the universality of the viscosity ratio for PTT model. The variations in uniaxial/planar elongational viscosities and the viscosity ratio with elongation rate are shown in Figs. 5 and 6 for the PTT model. The relaxation time and nonlinear material parameters in the models were again chosen to involve strong (Model A fluid) and weak (Models B and C) extension-thickening properties. It is found that Model C fluid with longer relaxation time shows the weaker extension-thickening but larger viscosity ratio for the most elongational rates. The computed film width for each model indicated that the Model C exhibiting the larger viscosity ratio showed the larger film width compared to Models A and B (Fig. 7), suggesting that the elongational viscosity ratio is useful not only for Larson model but also for PTT model for describing the film deformation in the casting process.

3.3 Cross Model

Next, we will show the variation in the elongational viscosity ratio and computed film width for Cross model. The Cross model exhibits particular shear-thinning properties but neither extension-thickening natures nor elasticity. The uniaxial and planar elongational viscosities for three different fluids are shown in Fig. 8. The Model A fluid exhibits a strong shear-thinning property in the range of elongation rate above 10. The Model B fluid involving a longer relaxation time is found to show the shear-thinning behavior in lower elongation rates. Figure 9 shows the ratio of uniaxial to planar elongational viscosity for these three fluids. Model A fluid exhibiting stronger shear-thinning shows the larger viscosity
It should be noted that the viscosity ratio in Fig. 9 is much closer to unity than that in Figs. 4 and 6 for the viscoelastic fluids, indicating that the Cross model exhibits the characteristics similar to those in Newtonian fluid. The calculated film width is shown in Fig. 10 plotted against the draw ratio. The film width monotonically decreases with increasing draw ratio because of the little extension-thickening properties of the fluid (cf. Figs. 3 and 7 for viscoelastic fluids involving extension-thickening natures). The solid curve represents the Model A fluid exhibiting a strong shear-thinning nature but high viscosity ratio. It is found from Fig. 10 that the Model A fluid shows the largest film width, while the Model C exhibiting the lowest viscosity ratio exhibits the smallest film width. These facts suggest that the ratio of uniaxial to planar elongational viscosity is universal in the film deformation not only for the viscoelastic fluids but also for the purely viscous fluid.

### 3.4 Comparison between Viscoelastic/Viscous Models

Figure 11 shows the ratios of uniaxial to planar elongational viscosity for the Larson, Cross and Newtonian models. Newtonian model exhibits a constant viscosity ratio of unity.
The viscosity ratio of any fluid models agrees with that of Newtonian fluid in low elongation rates. It is found from Fig. 11 that the viscosity ratios of Cross models are ranging between the Larson and Newtonian models for any elongation rates. The computed film width in Fig. 12 indicates that the predicted film widths agree with the Newtonian solution for draw ratios less than 4, corresponding to the constant viscosity ratio of unity in the low elongation rate regimes. Figure 12 demonstrates that the film width of Cross models is ranging between the Larson and Newtonian models as is the viscosity ratio, suggesting again that the viscosity ratio is the key rheological property that dominates the film deformation in the film casting process. Here it is interesting to note that fluids exhibiting a viscosity ratio of unity can involve the same deformation kinetics as those for Newtonian fluid. Sakaki\(^3\) showed that the Newtonian solution involving no elasticity was in good agreement with the experimental data for viscoelastic HDPE. Because the film width is controlled by the viscosity ratio as we proposed here, this peculiar agreement between the experiments and simulation might be due to the fact that the HDPE exhibits the viscosity ratio of unity for a wide range of elongation rates. Direct measurements of the planar elongational viscosity of HDPE would provide us detailed physical descriptions for the mechanism of film casting process, although few experimental data are currently available.

### 4. CONCLUSIONS

The quasi-three-dimensional simulation of film casting was performed for isothermal two viscoelastic fluids using Larson and PTT models, and purely-viscous non-Newtonian fluid using Cross model and Newtonian fluids. The results indicated that the film width was determined by the ratio of uniaxial to planar elongational viscosity rather than the extension-thickening nature in uniaxial elongation for both viscous and viscoelastic fluids. This fact suggests the neck-in phenomenon arises from a subtle balance of the uniaxial/planar elongational deformations, depending upon the rheological properties of the fluids.

### REFERENCES