Theoretical and Experimental Analysis of Interfacial Instabilities in Coextrusion Flows

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ABSTRACT: Interfacial instabilities developed during two-layer flows of LDPE melts were investigated theoretically through viscoelastic FEM analysis as well as experimentally on a flat multi-manifold coextrusion die. During continuous reduction of the minor layer, the wave type appears in the film first whereas the zig-zag type is only visible later, at the much thinner minor layer, which is an opposite order compared to film blowing coextrusion. Moreover, extensional viscosity of the minor layer was found to play a significant role from the interfacial instability point of view. © 2005 Wiley Periodicals, Inc. J Appl Polym Sci 98: 153–162, 2005

Key words: coextrusion; modeling; interfaces; polyethylene

INTRODUCTION

Increasing demands on the properties of plastic products have supported the development of coextrusion technology, in which two or more layers are extruded through the same die. The result then is a continuous film, sheet, tube, or profile that combines the properties of the extruded materials.

However, there are still flow phenomena that are not fully understood yet even if they represent the main limiting factor in the coextrusion technology. Interfacial instabilities are one of them. They represent an internal type of instability (inside the product), that is, the outside surfaces may be smooth in this case (Fig. 1). To avoid these internal defects, producers pay attention to the identification of conditions under which the instabilities appear in practice, and this is also researched on the theoretical level.

Intensive research of interfacial instabilities started in the late 1970s by Shrenk and coworkers[1] and Han and Shetty,[2] who studied the coextrusion flow of materials with different properties. They suggested critical interfacial stress as a criterion for the onset of interfacial instabilities. Their followers, Mavridis and Shroff,[3] moved the research a step further and employed computer simulations based on a discovery that the key to smooth parallel flow of materials is minimizing the interfacial shear stress and matching the elastic properties of adjacent layers.

Ramanathan and coworkers[4,5] and Perdikoulias[6] continued the investigations and distinguished two basic types of interfacial instabilities—zig-zag and wave. The former are small (small amplitude) with high frequency, while the latter have low frequency and are much bigger (large amplitude). The zig-zag instabilities were proved to be controlled by critical shear stress (Schrenk's theory); however, this parameter was found to be influenced by a number of factors, including among others arrangement and thickness of layers.[2] The origin of wave instabilities was considered to be more complex.

Wave type instabilities reported in refs. 6–16 have been found to be linked to elongational properties of the materials and extreme deformation of the layers at the merge point, that is, the point where the coextruded layers meet. It was also proved by Perdikoulias[6] for annular dies and Martyn and coworkers[10–15] for flat dies that instabilities can appear even for the same materials in adjacent layers at a certain ratio of thicknesses.

For the description and modeling of material flow, various constitutive equations are employed in FEM analysis. Good results have lately been reached with the modified White–Metzner model[17] and the modified Leonov model,[18] as reported in refs. 16 and 19–21. This model enables precise description of viscoelastic behavior, it has a capability to capture both hardening and softening phenomena in simple extension of LDPE, and from the mathematical point of view it is stable.

Even if the role of material properties and processing conditions in the appearance of interfacial instabilities has been quite intensively studied, both theo-

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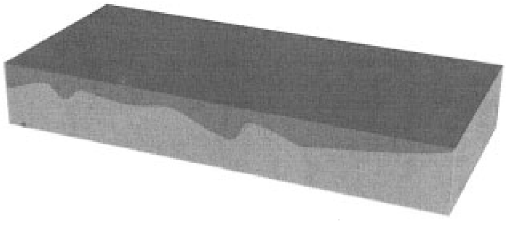

Figure 1 Scheme of interfacial instabilities at coextrusion.

retically and experimentally, there are still a number of unanswered questions. Theory and practice are closely linked here; obtaining theoretical background of the problem will help in selecting suitable materials and conditions for the coextrusion process, and the other way around, the data from experiments will enable making mathematical description of the process more accurate.

Keeping this in mind, in the present article, we analyze instabilities in the coextrusion process. First, the flow behavior of the layers is modeled and then the conclusions are verified in experiments. In both, the role of extensional viscosity on interfacial instabilities is followed. Moreover, experimental monitoring of interfacial instabilities in a flat die is compared with those observed in the film blowing process by Perdikoulias.[6]

Note that special attention is paid here to the investigation of whether theoretical conclusions from ref. 8 are in good agreement with experimental reality and if they are also valid for coextrusion flows with mixed shear and planar extensional deformation rate components appearing in flat dies. The newly proposed Leonov model[18] will be employed and tested for this purpose.

EXPERIMENTAL

For the study, LDPE materials commonly used in flat die coextrusion for film production were chosen, or more precisely, two types of polymers, two different lots for each of them. The notation of the materials in the following is LDPE 1/1, LDPE 1/2, LDPE 2/1, and LDPE 2/2 (i.e., polymer grade/lot). In the theoretical part of this paper, the modeling of the flow behavior will be described on LDPE 1/1 and LDPE 1/2; the experimental part employs LDPE 2/1 and LDPE 2/2. This choice of different polymers for the modeling and experimental parts has the following background. The two lots chosen for the theoretical part have the highest differences in extensional viscosities from all investigated grade/lots. Therefore, for these two materials (LDPE 1/1 and LDPE 1/2), the highest differences in stability of the coextrusion flows can be theoretically demonstrated as the extreme (limiting) case, which may occur during coextrusion of different lots. On the other hand, LDPE 2/1 and LDPE 2/2 were used in the experimental part because their elongational viscosity differences were found to be typical of all investigated grade/lots.

The materials were characterized by molecular weight distributions (MWDs), determined through High Temperature Gel Permeation Chromatography using a differential refractive index detector, relaxation spectra, and shear and extensional viscosities. The results for individual materials are presented in the corresponding part of the article.

For rheological measurements, two devices were used: low-shear-rate viscosity data were determined on an ARES (Rheometrics Scientific) parallel-plate rheometer, whereas for high-shear-rate viscosity measurements, an RH7–2 capillary rheometer (Rosand, UK) was used. The same instrument served for the determination of uniaxial elongational viscosity from the measured entrance pressure drop. The exact procedure is described further.

Relaxation spectra of the materials were determined by fitting the loss and storage moduli versus frequency data by the Maxwell model.

RESULTS AND DISCUSSION

Modeling of coextrusion flow

Materials characterization—structure and rheology

As said above, modeling was performed for two branches of LDPE 1. Their MWDs and polydispersities are depicted in Figure 2 and Table I. The materials'

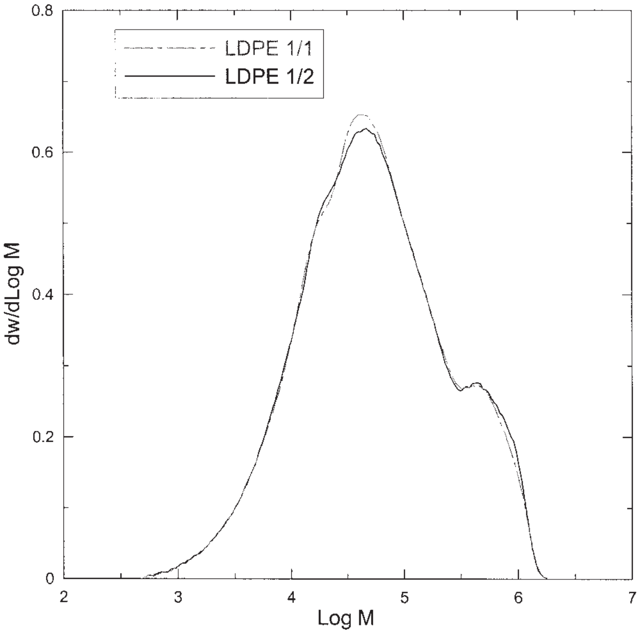

Figure 2 Molecular weight distributions of LDPE 1/1 and LDPE 1/2.

TABLE I
Materials Polydispersities

Material	M_w/M_n	M_z/M_n
LDPE 1/1	8.621	67.07
LDPE 1/2	6.502	43.92

MWDs are similar, but differences can be spotted in some parts. As can be seen, the lots differ in both M_w/M_n and M_z/M_n, the latter difference being larger.

Uniaxial elongational viscosity was calculated from the measured entrance pressure drop (capillary rheometer) in the following way, proposed by Zatloukal and coworkers:[22] First, apparent entrance viscosity, η_{ENT}, was determined from the entrance pressure drop (measured on a zero-length die), P_0, and apparent shear rate, $\dot{\gamma}_a$.

$$\eta_{\mathrm{ENT}} = \frac{P_0}{\dot{\gamma}_a} \tag{1}$$

Then, the apparent entrance viscosity was fitted by eq (2), where the plateau value, $\eta_{ENT,0}$, at low shear rates is known from Newtonian viscosity:

$$\log(\eta_{\mathrm{ENT}}) = \log\left(\frac{\eta_{\mathrm{ENT},0}}{1 + (\lambda\dot{\gamma}_a)^a}\right)\left[\frac{\tanh(\alpha\dot{\gamma}_a + 1)}{\tanh(1)}\right]^{\xi} \tag{2}$$

This is an empirical equation proposed in ref. 22, which combines the Cross model and an additional term that allows a maximum to appear in the entrance viscosity. Parameters α and ξ control the shape of the entrance viscosity maximum. Equation (2) is used for the prediction of viscosity values at very low shear rates where the measurements are impossible, and this data is taken for experimental values in further calculations. The parameters of the equation for the investigated materials are summarized in Table II, and the comparison between the two measured entrance viscosities, along with their fitting lines, are given in Figure 3. As can be seen, the viscosities differ mainly in the low apparent shear rate region.

The final step in elongational viscosity determination was the use of the Cogswell[23] and Binding[24] methods for the determination of extensional viscosity in the strain hardening and softening parts of the flow curve, respectively, and "Effective Entry Length Correction"[22] was applied to deal with all extensional viscosity data. Two different techniques were employed to determine the extensional viscosity curve because the Cogswell method was shown in ref. 22 to be better in low extensional rates, whereas the Binding method was better for high extensional rates after applied correction. The obtained extensional and shear viscosities of both lots of the same polymer are presented in Figure 4. An important point here is the nearly identical shear viscosity and significantly different elongational viscosity for various lots of the same polymer. As a result, the samples behave in a different way when exposed to extensional flows, which are present in both extrusion dies (converging and diverging sections, merging area in coextrusion) and postdie processes (film blowing, film casting, fiber spinning, etc.). On the other hand, in simple shear flows, the behavior of both samples will be practically identical.

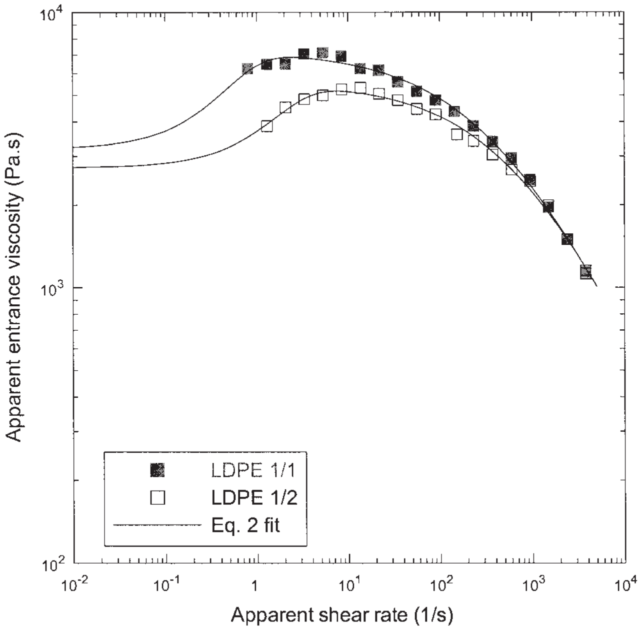

Figure 3 Apparent entrance viscosities for two different lots of LDPE 1, 210°C.

The analysis of molecular weight distribution (Fig. 2) shows that both lots have a similar M_w/M_n ratio. This is reflected in the materials shear flow properties (Fig. 4); the curves are nearly identical with only minute differences in the Newtonian plateau. On the other hand, the M_z/M_n ratio differs much more significantly, which is reflected in a different level of

TABLE II
Parameters in eq. (2) for the Materials Melts, 210°C

Material	$\eta_{\mathrm{ENT},0}$ (Pa s)	λ (s)	a	α (s)	ξ
LDPE 1/1	3187.0	0.002600	0.624843	1.095224	0.353630
LDPE 1/2	2731.5	0.001511	0.653400	0.306302	0.312904

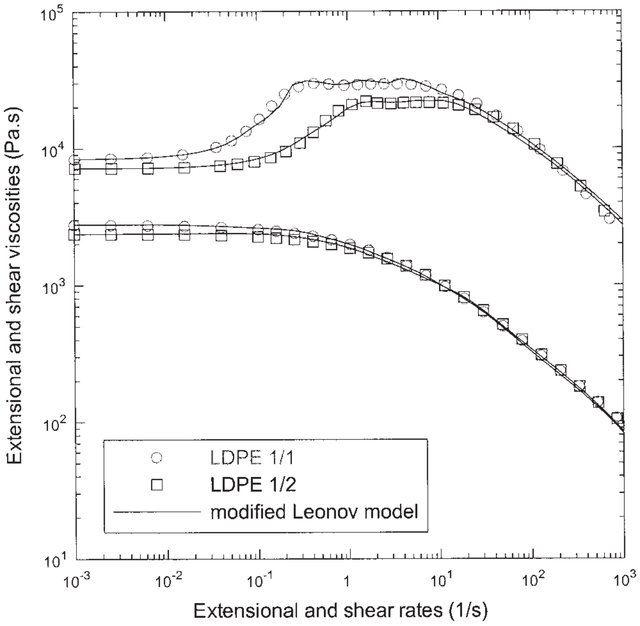

Figure 4 Extensional and shear viscosities for two different lots of LDPE 1, 210°C.

strain hardening in the extensional viscosities of the lots. This indicates that the LDPE 1/1 has a higher level of long chain branching than LDPE 1/2.[25]

Employed constitutive equations

For the flow simulation, the following models were used:

Modified Leonov model, which is based on heuristic thermodynamic arguments resulting from the theory of rubber elasticity.[26,27] In the model, the fading memory of the melts is introduced through an irreversible dissipation process. The dissipation term, *b*, is in the following form:[18]

$$b(I_1) = \frac{1}{4\lambda}\left\{\exp(-\,\xi\sqrt{I_1 - 3}) + \frac{\sinh[v(I_1 - 3)]}{v(I_1 - 3) + 1}\right\} \quad (3)$$

The parameters of the modified Leonov model are the relaxation spectrum and adjustable parameters ξ and ν, which can vary with relaxation time; they are presented in Table III. It should be pointed out that the adjustable parameters ξ and ν have been determined using the steady uniaxial extensional viscosity data only. Figure 4 proves that the mathematical description of the flow by this model is very precise.

Modified White–Metzner model, mWM, was the second model used in the simulation. It is basically the Maxwell model, in which the viscosity and relaxation time are allowed to vary with the second invariant of the strain rate tensor. The extensional viscosity is forbidden to yield infinite extensional viscosity[17] by the following functions:

$$\eta(II_d) = \frac{\eta_0}{[1 + (K_1\sqrt{2II_d})^a]^{1-n/a}}, \quad (4)$$

$$\lambda(II_d) = \frac{\lambda_0}{1 + K_2 II_d} \quad (5)$$

with

$$\frac{\lambda_0}{K_2} < \frac{\sqrt{3}}{2} \quad (6)$$

where η_o means the Newtonian viscosity, and λ_0, K_1, K_2, n, and a are constants. The parameters of the model are given in Table IV.

Analysis of coextrusion flow stability

The flow analysis of coextrusion from the point of view of stability was performed through a fully viscoelastic FEM analysis (details in ref. 19), together with the recently proposed Total Normal Stress Difference (TNSD) sign criterion [7,8,20], which characterizes the relative stretching of the coextruded layers in the merging area and was found to be useful for

TABLE III
Model Parameters for LDPE 1/1 and LDPE 1/2 Melts, 210°C

	LDPE 1/1				LDPE 1/2			
	Maxwell parameters		m Leonov model parameters		Maxwell parameters		m Leonov model parameters	
i	λ_i (s)	$G_{0,i}$ (Pa)	ξ	ν	λ_i (s)	$G_{0,i}$ (Pa)	ξ	ν
1	0.00114	80412.4	0	0.2	0.00116	82290	0	0.2
2	0.005	16133.4	0	0.2	0.00563	18021.5	0.5	0.2
3	0.02202	11565.2	0.8	0.04	0.02733	10808.3	0.95	0.04
4	0.09694	4312.37	0.8	0.014	0.13254	3774.5	0.5	0.014
5	0.42675	1585.79	0.6	0.014	0.64286	1136.01	0.5	0.014
6	1.87872	385.162	0.7	0.01	3.11791	200.183	0.3	0.01
7	8.27079	36.5673	0.25	0.01	15.1222	1.85962	0.25	0.01
8	36.4100	6.13736	0.2	0.001	-	-	-	-

TABLE IV
Parameters of Modified White–Metzner Model

	η_0 (Pa s)	λ_0 (s)	K_1 (s)	K_2 (s)	n	a
LDPE 1/1	2771.3	8.6042	0.3044	10.518	0.4154	0.6662
LDPE 1/2	2375.2	2.3403	0.1991	2.8496	0.3952	0.7067

determination of the onset of the wave type of instabilities. In more detail, it was revealed that the wave instabilities appear at the interface if TNSD changes its sign in the merging area or is negative. In these cases, an elastic after−effect or minor layer breakdown occurs. This was suggested to be the physical reason for the onset of the wave type of instabilities. Moreover, the extensional viscosity was shown to be the driving parameter in this case.[8,20] Thus, we concentrate here on the wave instabilities because the chosen polymers have practically identical shear viscosities and different extensional ones. This allows us to investigate the direct effect of the extensional viscosity on the stability of the interface.

Simulation analyses were performed on a standard flat multi-manifold coextrusion die, described in detail in the Experimental section. The details of the merging area of the die are depicted in Figure 5. Two combinations of two different lots of the same polymer grade were investigated in the simulation of a two-layer flow with the help of the TNSD sign criterion. The total mass flow rate was kept constant, and the thickness of the minor layer was changed. (In the following, the percentage always means the share of the minor layer in the total mass flow.) The FEM analysis of coextrusion was performed by the FLOW2000™, 2D FEM module.[28] In the flow modeling, the velocity profile and the interface location were obtained through the viscoelastic modified White–Metzner model, while the TNSD values were calculated by the modified Leonov model.

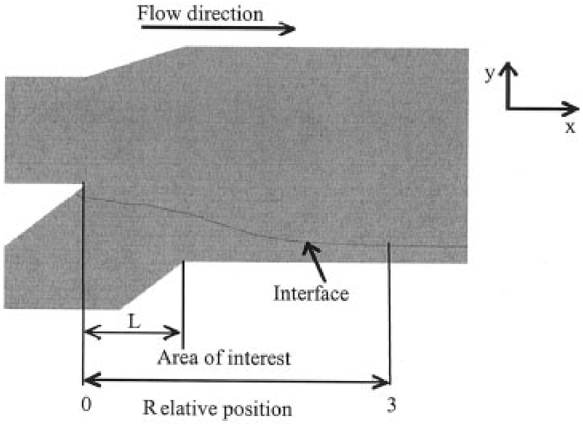

Figure 5 Merging area of the flat coextrusion die.

First, an analysis of coextrusion stability was performed for different lots in both coextruded layers. This was done through the TNSD sign criterion,[7,8,20] and the results are depicted in Figure 6. To describe the area of interest, the relative position was labeled x_c/L, where x_c represents the position on the x coordinate axis (x_c is equal to zero at the merging point) and L is given in Figure 5. The area of velocity rearrangement that is studied corresponds with the relative position $\langle 0, 3\rangle$.

The TNSD sign criterion (Fig. 6) proves very clearly that for the coextrusion with the same polymer grade in both layers, the stability of the flow is influenced by the choice of the lot for a particular layer. More precisely, if LDPE 1/1 (high extensional viscosity) and LDPE 1/2 (low extensional viscosity) are used in the minor and major layer, respectively, the coextrusion is wavy unstable. However, when the lots are switched, the flow is stable.

Another approach to eliminating interfacial instabilities is increasing the mass flow rate in the minor layer. The result in Figure 6 shows that for this combination the flow becomes reasonably stable at 10% of the total mass flow rate in the minor layer. On the other hand,

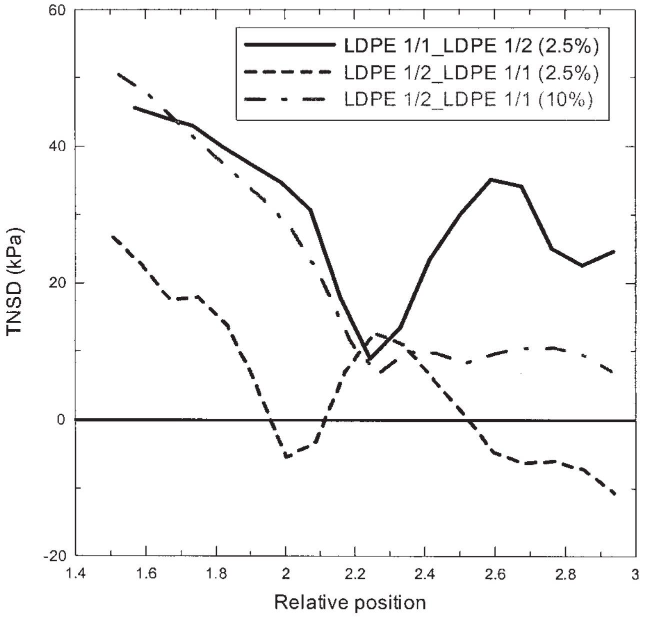

Figure 6 The course of TNDS functions along the interface in the merging area, different lots of the same polymer (LDPE 1), different mass flow rates in the minor layer.

TABLE V
Materials Polydispersities

Material	M_w/M_n	M_z/M_n
LDPE 2/1	4.255	20.16
LDPE 2/2	4.137	15.48

the same stability was achieved for much thinner layers (2.5%) when the lots in layers were swapped. This clearly shows how important a role is played by the variation in the flow properties of various lots, even if they are declared as the same material by the producer. Quantitatively, the use of different lots of the same polymer in our study (having pronounced differences in extensional viscosity) shifted the stability of the process between 2.5 and 10% of the minor layer share.

A simple conclusion could be drawn from this theoretical result: For materials having similar shear viscosities, the flow is more stable when the minor layer material has lower elongational viscosity. The materials comparison shows that the elongational viscosity changes with the lot even if the shear viscosity is almost identical. Therefore, the process may be stable when the lot with lower elongational viscosity is in the minor layer, and can become unstable when the elongational viscosity of the minor layer is higher.

This result can also explain a very often observed phenomenon from the polymer industry—that when a process is running stable for a certain time and then, without any observable changes, it becomes unstable and after a while it stabilizes again. The reason is that the material elongation properties are changing over the time when different lots of the same grade are used for the production.

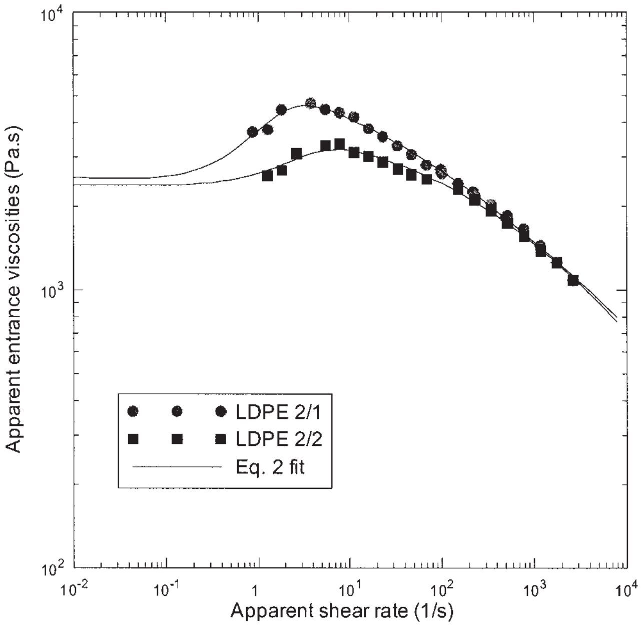

Figure 7 Entrance viscosities of LDPE 2.

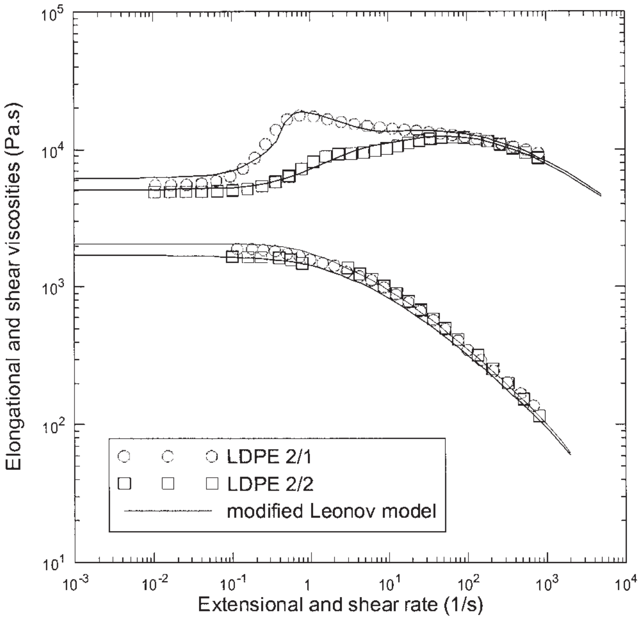

Figure 8 Elongational and shear viscosities of LDPE 2.

Experiments

As in the previous part, two different lots of the same LDPE grade were used for the experiments, LDPE 2/1 and LDPE 2/2. The criterion for the selection of materials was to obtain the M_w/M_n ratio very similar to the material used by Perdikoulias[6] in film blowing, so that we could compare interfacial instability development in the flat (this work) and annular coextrusion dies.[6]

The molecular characteristics of LDPE 2 are given in Table V. As can be seen, both lots have very similar M_w/M_n but they differ in M_z/M_n. The rheological properties of the materials are presented in Figures 7 and 8. As in the modeling part, here also the entrance viscosity and determined elongational viscosity was fitted by eq. (2) and the modified Leonov model, respectively. The model parameters are given in Tables VI and VII. As can be seen, also in this case both lots of LDPE 2 have nearly the same shear viscosities, while the elongational viscosities in the region of our

TABLE VI
Parameters in Eq. (2) for the Materials Melts, 230°C

Material	$\eta_{ENT,0}$ (Pa s)	λ (s)	a	α (s)	ξ
LDPE 2/1	2700	0.03373	0.3290379476	0.489019	0.448548
LDPE 2/2	2420	0.002906	0.4160167773	0.206259	0.228092

TABLE VII
Model Parameters for LDPE 2/1 and LDPE 2/2 Melts, 230°C

	LDPE 2/1				LDPE 2/2			
	Maxwell parameters		m Leonov model parameters		Maxwell parameters		m Leonov model parameters	
i	λ_i (s)	$G_{0,i}$ (Pa)	ζ	ν	λ_i (s)	$G_{0,i}$ (Pa)	ξ	ν
1	0.000594317	105250	0.12	0.003	0.00049	105240	0.2	0.003
2	0.00265157	26400.50	0.12	0.003	0.0019	21987.7	0.2	0.003
3	0.011783485	18229.2	0.12	0.003	0.00741	19605.4	0.2	0.003
4	0.052442792	7259.84	0.12	0.003	0.02892	8129.94	0.2	0.003
5	0.233366712	2575	0.14	0.003	0.11284	3541.64	0.19	0.003
6	1.038432436	510.29	0.62	0.009	0.44037	1024.71	0.24	0.009
7	4.620754614	31.8944	0.4	0.009	1.71852	149.089	0.22	0.009
8	20.56120911	2.66116	0	0.1	6.7064	6.9614	0	0.1
9	-	-	-	-	26.1711	2.41581	0	0.1

interest differ quite markedly as a result of different levels of long chain branching, as discussed in ref. 25. Also in this case, the modified Leonov model has good capability to describe the rheology of both lots (Fig. 8).

For the materials with determined flow properties, the conclusions about instabilities obtained in the modeling part were verified by coextrusion experiments performed on a commercial coextrusion pilot plant line. A standard flat multi-manifold coextrusion die 300 mm wide was used, whose geometry is depicted in Figure 9. For the measurements, the minor layer was colored with carbon black. The total output mass flow rate was maintained constant (24kg/h) throughout the test, and that in the minor channel was gradually reduced. After any change, the process was allowed to steady, and then visual assessment of the coextruded film was done.

The onset of wave type instabilities

The results of the experiments are presented in Table VIII (onset of instabilities), and the appearance of instabilities for LDPE 2/1 is documented in the photos in Figure 10.

As the minor layer seems to have high importance, we first used exactly the same material (LDPE 2/1—the same lot) in both layers and changed the thickness of this layer. Surprisingly enough, even for identical materials in both layers, the produced film showed quite clear instabilities. For a thin minor layer, the film suffers from both zig-zag defects and waves; however, the difference is in the point of onset of the particular type of instabilities. This confirms conclusions from refs. 4 and 5 that the origin of each instability is different. This experiment also proved that both instabilities are present even when identical materials are in both layers and, therefore, the instabilities cannot only be attributed to material differences. The flow history seems to play a significant role here, as suggested in ref. 7.

In the second step, the original material (LDPE 2/1) in the minor layer was replaced by LDPE 2/2, that is, by the material with lower elongational viscosity. As can be seen from the Table VIII, waves show quite an important shift—the flow is only weakly unstable for the minor layer of 4.2%. This confirms the findings from modeling that lower elongational viscosity supports the stability of flow from the viewpoint of waves.

To be more precise, the theoretical analysis based on the TNSD sign criterion was also employed here, with

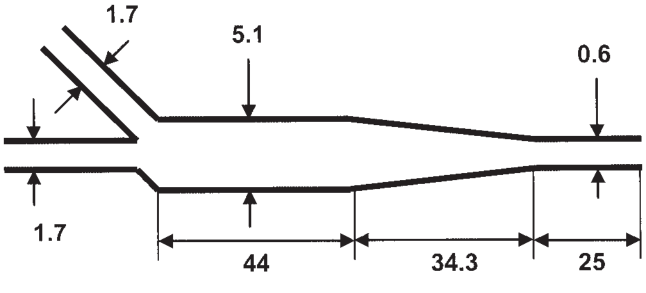

Figure 9 The die geometry.

TABLE VIII
Wave Instabilities for Various Lots in the Layers (s - strong, w - weak)

Minor layer mass flow rate (%)	LDPE 2/1 in both layers	LDPE 2/1 - major LDPE 2/2 - minor
	Wave instabilities	
2.9	Unstable - s	Unstable - s
4.2	Unstable - s	Unstable - w
5.4	Unstable - s	Unstable - w
8.3	Unstable - w	Stable
13.8	Stable	Stable

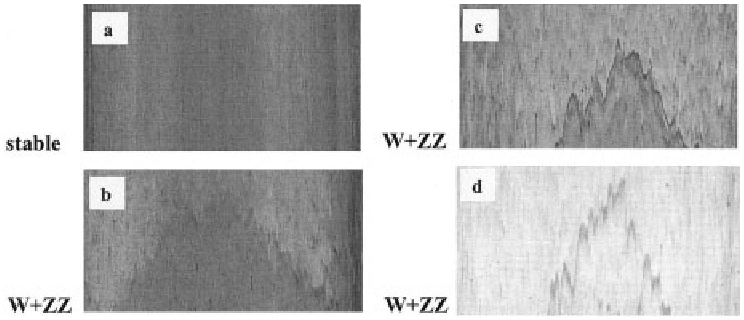

Figure 10 Wave (W) and zig-zag (ZZ) interfacial instabilities in coextruded films with LDPE 2/1 in both layers. Minor layer mass flow rate: (a) 13.8%, (b) 8.3%, (c) 5.4%, and (d) 4.2%.

the aim to correlate the experimental findings in this section. First, for the combination of 2/1 and 2/1, the TNSD method predicts the instability onset for 7% thickness of the minor layer (Fig. 11). This is in good agreement with the results shown in Figure 10 and Table VIII (8.3%). Second, it can be seen from experiments, summarized in Table VIII, that the system is more stable when lot 2/2 is used in the minor layer (2/1 and 2/2 structure) compared to 2/1 and 2/1 structure. This corresponds with the calculation results from Figure 11, where the former structure is also predicted to be more stable. These findings show that the trends predicted by the TNSD sign criterion are in a good correspondence with the experimental results.

The development of interfacial instabilities

The development of interfacial instabilities in a flat coextrusion die was investigated through continual reduction of the minor layer thickness at constant output mass flow rate. Figure 12 shows the general trend of this process for LDPEs. It should be said that Figure 12 is meant as a representative picture composed of several samples and process conditions just to show all the stages of instability development. We actually did not observe all these stages in any experiment with two materials when the layer share was changed. More detailed information about the experimental work can be found in ref. 29.

When the minor layer is thick enough, the flow is stable (first snap). As the mass flow rate in the minor layer decreases, wave instabilities start to appear in the film. They are first weak and poorly developed, but when the minor layer is still thinner, their contours become sharper and better visible (W1). A further reduction of the layer causes the development of other waves inside the original one; the wave multiplies and penetrates the film thickness (W2). Then, zig-zag in-

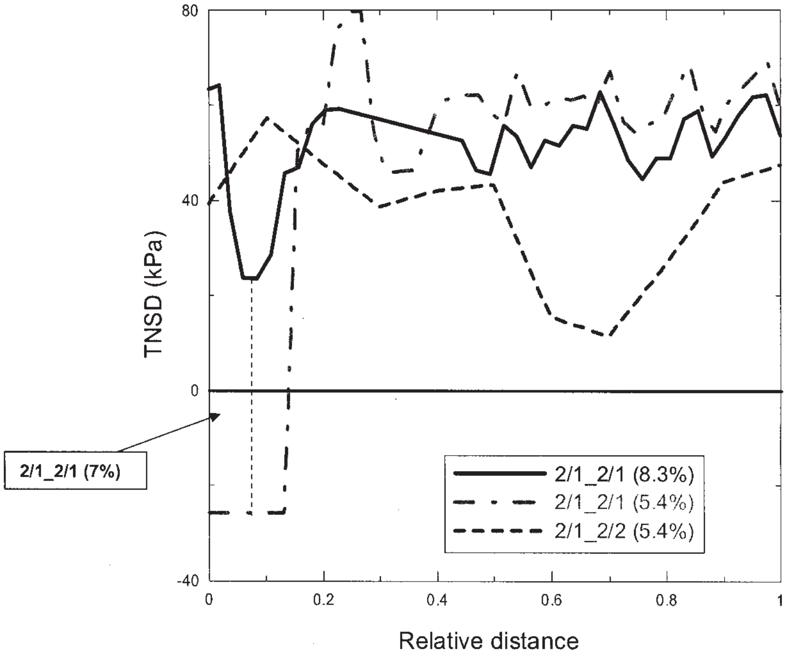

Figure 11 The course of TNDS functions along the interface in the merging area, different lots of the same polymer (LDPE 1), different minor mass flow rates. Dashed line represents interpolation line between minimum TNSD values for 8.3% and 5.4%.

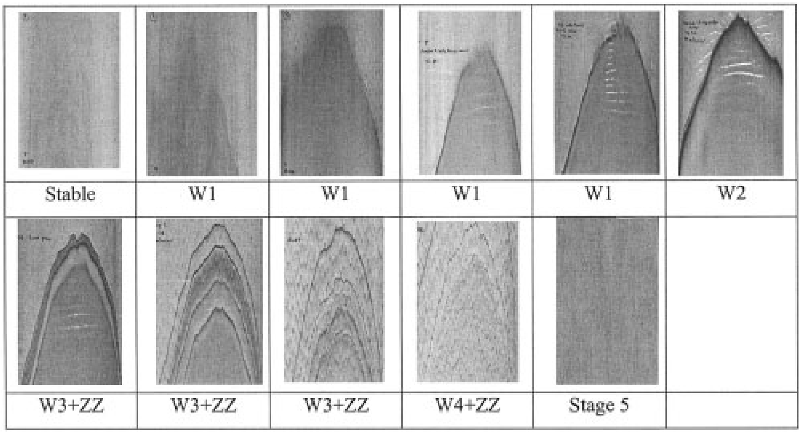

Figure 12 Development of LDPE interfacial instabilities with minor layer being reduced (W –wave, ZZ –zig-zag instabilities).

stabilities appear gradually in the whole film area (W3+ZZ). With a minimized minor layer, the wave instabilities become significantly destroyed (W4+ZZ). Finally, for a very thin minor layer, the flow becomes "stable" again (Stage 5). The reason for this is probably that there is practically no material in the layer and, instead of coextrusion, a single-layer extrusion starts.

From the results in this section we can see that waves start before zig-zag instabilities, for a thicker layer. In other words, zig-zag instabilities start at more severe conditions. Thus, from the production point of view, zig-zag instabilities are not so important because the product is damaged as soon as the waves start.

Development of interfacial instabilities in different technologies

Another point of view of the assessment of interfacial instabilities is the technology where they appear. To compare the technologies of cast extrusion and film blowing, we will use the results of our research for cast film and results published in ref. 6 for film blowing.

As proved earlier,[1,6] an important role in the onset of instabilities is played by the critical interfacial shear stress. It is calculated from the process conditions and the materials rheology. When the shear stress at the interface exceeds the critical value, zig-zag instabilities appear. From this point of view, we wanted to compare the behavior of very similar materials in different processes—film blowing and cast extrusion, that is, to compare our results with those presented by Perdikoulias.[6] For the experiments, materials with very much the same M_w/M_n for cast (LDPE 2/1, LDPE 2/2) and blowing (LDPE NA 345–009) extrusion were chosen (M_w/M_n = ca 4). The critical shear stresses determined in our research are given in Table IX. Compared to the values obtained in ref. 6 for film blowing, which were 65 to 72 kPa, our results are very similar (66–67 kPa). This indicates that one critical interfacial shear stress may exist for LDPEs having similar M_w/M_n, even for different coextrusion technologies. On the other hand, the compared materials had similar polydispersity characteristics. Therefore, we cannot conclude from these two measurements that the value for the critical shear stress would be the same for different material MWDs. This should be another topic for investigation.

Differences, however, can be seen in the type of interfacial instabilities. In the flat die (cast film), the temperature of the material is higher (230–250°C), that is, the shear viscosity is lower, which results in lower shear stresses. That is why the critical shear stress is hardly reached at the end of the die and the first type of interfacial instabilities to appear is waves. In blow die, on the other hand, the temperatures are much lower (about 190°C), which causes higher shear stresses, so the critical shear stress is easily reached and zig-zag instabilities are set before the waves as shown in ref. 6.

CONCLUSIONS

The analysis of flow instabilities during extrusion has shown some interesting features of the process:

Modeling revealed that elongational viscosity affects flow instabilities in flat coextrusion dies

TABLE IX
Critical Shear Stresses for Combinations of LDPE 2 Lots

Coextruded materials	Shear stress stable (kPa)	Shear stress unstable (kPa)
LDPE 2/1 & LDPE 2/1	61	67
LDPE 2/1 & LDPE 2/2	60	66

(higher η_E of the melt in the minor layer than in the major one, more susceptible interface to wave type of interfacial instabilities). This was also proved by experiments.

Changing lots in the minor layer may affect flow stability. This indicates that material rheology, and mainly elongation properties, are the key parameter for understanding and predicting interfacial instabilities. Therefore, for any quantification of these instabilities, both shear and elongational viscosities must be taken into account.

The work suggests that in coextrusion, one critical interfacial shear stress may exist for LDPEs having similar M_w/M_n ratio.

The mapping of interfacial instabilities during film casting revealed that at a continuously reduced minor layer, the wave type appears first. The zig-zag type is only visible later, at a much thinner minor layer.

The TNSD sign criterion was found to be a useful tool for the evaluation of the wave type of interfacial instabilities during coextrusion flows in a flat die.

The modified Leonov model was proved to have high capability to describe extensional rheology of LDPE melts commonly used in the cast film process.

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