Correcting Flow Instability in Coextrusion

Flow instabilities such as interfacial instability, melt fracture, or layer non uniformity are common problems that could cause quality problems in coextrusion. There are several options to correct these problems, but the key is to select the best option or combination of options to eliminate the problem and minimize other possible adverse effects. Flow instabilities in coextrusion can cause quality problems or limit production rates. Appropriate operating conditions of the coextrusion process combined with proper polymer selection can successfully be used to resolve these troublesome quality problems.

There are several types of flow instabilities that can occur in the coextrusion process that cause non-uniformity problems to occur. Some of these instabilities include:

1. Extruder Instability (melt uniformity)
   - Pressure - varies with time.
   - Temperature - varies with time and location.

2. Steady State Instability (layer uniformity)
   - Polymer viscosity mismatch
   - Poor die design or assembly

3. Non-Steady State Instability (Elastic Instability)
   - Melt Fracture
   - Interfacial instability
   - Zig-Zag
   - Wave
   - Die swell

Melt Uniformity
Extruder surging is measured by variations in output rate with time. Surging is normally also associated with variations of pressure in the adapter. Good pressure variation for a single screw extruder should be less than +/- 1% (in the adapter pipe). Melt temperature can be measured by a variable depth thermocouple located in the adapter. The average melt temperature can be controlled by adjusting the barrel profile to achieve the desired temperature. The temperature difference across the adapter pipe should be minimized to achieve good melt uniformity.

Gravimetric feed control will assure that the average layer ratio targets are achieved. Good screw designs should be selected to provide good melt uniformity at the target output rates. The availability of good instrumentation and documentation is critical in determining if an extruder is experiencing a surging or poor melt uniformity problems.

Steady State Instability
A second type of instability can be described as steady state instability. The steady state instability means the polymer flows will reach some equilibrium condition, but the film does not have a uniform distribution of the layers. The non-uniform layer distribution can be caused either by a mismatch of polymer viscosity, or by the design of the coextrusion die. The importance of viscosity in maintaining layer uniformity has been studied extensively. The higher viscosity will tend to flow to the path of least resistance, and the lower viscosity polymer will tend to encapsulate.
Layer non-uniformity is particularly prevalent in flat dies because the lower viscosity polymer layer is pushed to the sides of the die. Annular dies tend to be slightly more tolerant of a viscosity mismatch. Some basic steps to solve non-uniform layer distribution problems include:

1. Increase the viscosity of the polymer layer that is spreading too thin. Lower the melt temperature of the polymer. Select a higher molecular weight polymer (lower Melt Index).
2. Decrease the viscosity of the polymer layer that is too thick. Raise the melt temperature of the polymer. Select a lower molecular weight polymer (higher Melt Index).
3. Adjust the die temperature profile (for flat dies).
4. Adjust the total output rates to achieve a better viscosity match.

(Some viscosity curves have a different sensitivity to shear rate) Sometimes the above methods help in resolving non-uniform layer distributions. In other cases, equipment design modifications might be required. For example, in feedblocks, feedslot shaping (when properly done) can correct a non-uniform layer distribution in flat dies. In blown film dies, cleaning and precise alignment the spiral mandrels is required.

Elastic Flow Instabilities
The non-steady state flow instabilities include melt fracture and layer interfacial instability. These instabilities are generally believed to be the result of a dynamic response of the elastic component of the polymer viscosity within the environment of the extrusion process. These elastic instabilities are often found to occur in the region of the highest shear stress.

These phenomenon might be described as folds or waves of polymer flow at an interface which results in a small periodic thickness variation in the polymer layer. The elastic interfacial instability will increase the roughness between the interface boundary.

Critical shear stress is a simplification of a complex response, but is many times used in describing elastic instability. The critical shear stress is defined as the shear stress at which the on-set of flow instability has been observed. The values reported in the literature can vary considerably for different polymer types and equipment design. The on-set of these instabilities occurs before the eye can detect the defects, furthermore, the point at which a defect becomes objectionable is also subjective.

Melt Fracture -
Melt fracture is instability at the polymer/metal interface on the film surface. Melt fracture is the folding of the polymer flow on the surface of the film. Again many theories on the nature of melt fracture have been proposed, but most agree that the elastic nature of the polymer is responsible for the instability. Melt fracture most often is reported to occur in the entrance of die land area, but some studies have seen melt fracture occurring at the exit of the die as well.

A key attribute relating to melt fracture is the sudden change in shear stress as in a contraction to high shear stress at the entrance of the die land or the sudden expansion to non-restrained flow at the exit of the die land. Birefringence measurements have confirmed that these are the locations of large stress concentrations in polymer flows. Using a critical shear stress, melt fracture has been reported for LLDPE polymers to occur from 1.0 to 2.3 x 106 dynes/cm2 (15 -33 psi).

Melt fracture is reduced by lowering the critical shear stress at the wall. Variables critical to reducing melt fracture are:
1. Viscosity in the skin layer could be reduced.
2. Output rate (total) could be reduced.
3. Die gap could be increased
4. Processing aids can be used to reduce the shear stress at the die surface.

Elastic Interfacial Instability -
Elastic interfacial instability or some times called Zig-Zag is a instability at a polymer/polymer interface and has been observed to occur in the die land. These instabilities may at low output rates, be nearly undetectable, and not interfere with the functionality of the coextruded film. If a large amplitude wave form develops at the interface
location, the polymer flow can cause intermixing at the interface boundary. When there are significant differences in the refractive index of the polymers the interfacial instability can more readily be seen. For example, with a HDPE and LLDPE film structure the HDPE is hazy enough to see the instability. Color pigments in a polymer will highlight interfacial instabilities. It is also possible to experience interfacial instability in coextrusion when running the same polymer in adjacent layers.

Few polymer coextrusion structures have been studied extensively enough to determine how the onset of instability varies with different polymer types. For some polymer systems the "critical shear stress" for interfacial instability is at 1/3 to 1/12 of the values reported for melt fracture. Interfacial instabilities have been reported at critical shear stresses of 0.5 to 0.8 x106 dynes/cm2 (6.3 -10 psi).

Wave Interfacial Instability -
Wave patterns (or sometimes called W 's) is instability at the polymer/polymer interface and occurs most often in asymmetrical structures. Wave instability has been found to occur where polymer flows converge with a large differences in velocities between polymer streams. These instabilities if they originate before the web is spread (i.e., in the feedblock) can be distributed over broad areas.

Die Swell Instability -
Die swell also has been reported to cause some instabilities. This occurs most frequently when one of the polymers has little die swell, while the adjacent polymer has high die swell. Here the instability begins at the die exit.

Reducing Interfacial Instability -
The steps to correct non-steady state instabilities detail how to reduce the shear stress at the interface in the coextruded structure, either at the polymer/polymer interface or the polymer/ metal interface. The steps include:

1. Output rates could be lowered.
2. Skin layer ratio could be increased.
3. Viscosity of adjacent layer could be reduced.
4. Die gap could be increased.

Decrease the total output rate -
This solution was not very economical as a long term solution, but it was successful as a short term solution and served to provide some empirical data. The lower output rate will change the slope of the shear stress profile, reducing the shear stress at the wall , and the shear stress at the polymer/polymer interface.

Increase the skin layer thickness
The highest shear stress is located at the wall of the die, increasing the skin layer ratio shifts the polymer/polymer interface toward the center of the flow channel, reducing the shear stress acting on the interface.

Decrease the viscosity of the skin layer polymer –
The next step was to determine if the viscosity could be lowered to reduce the shear stress at the wall. The viscosity can be decreased by raising the temperature of the polymer or by using a resin with a lower viscosity (higher melt index). A lower viscosity will shift the slope of the shear stress profile, thus lowering the shear stress at the wall.

It can readily be seen from this discussion that the term "viscosity matching" for coextrusion is not always for the best method. There will be some in stances where it is best to deliberately mismatch viscosity of the polymers to allow successful fabrication of the desired film structure.

Increase the die gap -
Another option to be considered is the die gap. This will change the geometry of the flow area, reducing the shear stress at the wall.

Summary
These methods can be used one-at-a-time or in combination to solve non-steady state instability problems. Though no complete predictive theory exists for these complicated rheological interactions, hopefully these guide lines along with some good empirical experience can help in troubleshooting coextrusion problems.

- Thomas I. Butler, The Dow Chemical Co.
See also:

- Causes of surging
- Coextruded sheets
- Defining screw performance
- Effect of temperature
- Flow surging
- Flow surging in single-screw, plasticating extruders
- Interfacial instabilities during coextrusion of LDPEs
- Melt temperature measurement
- Pyrometers
- Thermocouple depth
- "Wave"pattern instability in multilayer coextrusion

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