THE EFFECTS OF PROCESSING CONDITIONS ON THE OPTICAL PROPERTIES OF POLYETHYLENE FILMS

Polyethylene (PE) is widely used in the production of film for the packaging of foods and other goods. The required properties of the film are specified in accordance with the application and customer preferences.

In applications where a high level of consumer appeal in the display of products is required, general packaging films are required to exhibit a range of optical properties. In such applications, the required optical properties include very low haze, very high gloss and very good transparency (clarity).

This paper outlines how the choice of resin and the processing conditions used to produce the PE film impact on its optical properties.

The appearance of a polyethylene (PE) film is associated with the way it reflects, scatters, absorbs and transmits light. Although the term ‘clarity’ is used loosely to describe film appearance, this is usually a subjective summation of various visual characteristics of the film. The properties that are usually measured include transmitted light scattering (haze), surface gloss and see-through clarity, though none of these (nor indeed all three collectively) completely defines the optical properties of a sample of film.

A range of polyethylene resins are used in the manufacture of packaging film. These resins include low density (LDPE), linear low density (LLDPE), metallocene linear low density (mLLDPE) and high density (HDPE) grades. In addition to the type of resin used to make the film, processing conditions also affect the optical characteristics of the final material.

ORIGIN OF OPTICAL IMPERFECTIONS

There are three basic causes of optical imperfections in polyethylene film:

- Surface irregularities caused by melt flow phenomena
- Crystallisation behaviour
- Melt and solid inhomogeneities

These irregularities are frozen into the film when it solidifies in the region of the freeze line or frost line.

Surface irregularities caused by complex elastic melt flow behaviour in the extrusion die tend to be most often associated with LDPE film.

Crystallisation defects are more often associated with film made from LLDPE, mLLDPE and HDPE. The molten extrudate of a narrow molecular weight distribution LLDPE will usually appear very clear on exiting the die, with haze potentially developing near the freeze line as the polymer crystallises. In addition, surface melt fracture, if not eliminated, will also lead to poor optical properties. HDPE film can tend to be quite opaque due to the high level of crystallisation.

Light scattering in the presence of melt or solid inhomogeneities such as fine filler particles (e.g. particles of antiblocking additive), grain, microgel, cross-linked particles and contamination is the third primary cause of optical imperfections.
EXTRUSION HAZE

The most important surface irregularities in LDPE type films are extrusion defects (termed ‘extrusion haze’) which originate from the complex elastic melt flow behaviour in the extrusion die (see Figure 1). This defect structure consists of small raised areas less than 5 mm in height. It is thought to be due to the buckling of the surface resulting from differential melt relaxation of the highly sheared melt.

![Extrusion haze 10µm](image)

**Figure 1. Basic types of surface irregularities in PE film – extrusion haze**

Because the defects arise from melt flow behaviour, the nature and magnitude depend upon the flow properties of the PE itself, and will also be influenced by any extrusion conditions which reduce the shear rate within the die. The biaxial drawing of the melt, as the bubble is blown, will decrease the height and extend the length and breadth of the defects, so leading to a change in the texture of the film surface. Hence the final texture will be dependent upon the degree of draw and blow ratio.

In addition, the melt has a chance to relax and the extrusion defects time to smooth out in the cooling zone up to the freeze line; any increase in the cooling time will decrease the magnitude of the defects finally frozen into the solid film.

The extrusion variables which influence the extrusion haze are:

- **Output rate (shear rate) through the die** – the extrusion defects will become more severe as the shear rate is increased.
- **Melt temperature** – a higher melt temperature allows better flow through the die and reduces the tendency for flow defects.
- **Freeze line distance** – a higher freeze line allows more relaxation time for the irregularities to be smoothed (see Figure 2).
- **Blow-up ratio** – an increase in the blow-up ratio will allow greater relaxation of the irregularities and a reduction in their size.
- **Haul-off rate (drawdown)** – higher line speeds increase the severity of the extrusion haze.
- **Die geometry** – wider die gaps reduce the shear rate in the die.

Extrusion haze also decreases as the die entry angle is widened.

![Freeze Line Distance](image)

**Figure 2. Characteristic variation of film haze with freeze line distance for LDPE film**

CRYSSTALLISATION HAZE

A major cause of surface roughness in PE films arises from the growth and aggregation of crystallites at, or near, the surface of the film (see Figure 3). The irregularities, termed ‘crystallisation haze’, are small mound-like features approximately 1 mm in size. They account for around 10% of the total haze in LDPE type films.

![Crystallisation haze 10µm](image)

**Figure 3. Basic types of surface irregularities in PE film – crystallisation haze**

In LLDPE type films, including mLLDPE, crystallisation-induced surface roughness is the major contributor to the haze, because of the ease in which larger crystallites and spherulites can grow compared with LDPE films. In many cases, LLDPE films can have rougher surfaces, and higher haze and poorer clarity than conventional LDPE films. The higher the density and Melt Flow Index (MFI) of the LLDPE polymer, the worse the optical properties will be.

HDPE has a much higher crystallinity than LDPE and LLDPE. As a result, HDPE films show a very high level of haze and are not suitable for applications where optical properties are important. However, in monolayer films, HDPE may be blended into LLDPE and LDPE at low ratios and still achieve adequate clarity. In coextruded films, if the HDPE is blended into the core layer, it is possible to achieve high clarity.

In addition to the PE properties, the magnitude and intensity of the crystallisation irregularities will also be significantly affected by the rates of nucleation and growth of crystallites during the cooling stage of the film blowing process. Crystallite size increases with longer cooling times and any change in
conditions that will extend the cooling time will therefore contribute to higher crystallisation haze. The important extrusion variables which influence the cooling time include:

- **Film haul-off speed** – an increase in line speed results in thinner film and a shorter travel time between die and freeze line i.e. quicker cooling, less crystallite formation and lower haze. With LLDPE, increasing the drawdown increases haze because of stress-induced crystallisation.

- **Blow-up ratio** – an increase in blow-up ratio for a given freeze line and haul-off speed will give longer cooling times and higher haze.

- **Freeze line distance** – high freeze lines lead to slower cooling, more crystallite formation and higher haze. This effect is particularly pronounced in LLDPE films (see Figure 4).

- **Output rate** – increased output rate for the same freeze line distance facilitates quicker cooling and therefore lower crystallisation haze.

- **Melt temperature** – any increase in melt temperature necessitates increased cooling; if the freeze line distance is maintained, then lower crystallisation haze will be obtained because of the higher cooling rates.

![Figure 4. Typical variation of haze with freeze line distance for LLDPE film, thickness 40 μm (MFI 0.5)](image)

Crystallisation effects also cause light scattering within the film itself, at the boundaries of spherulites and between crystallites. This internal scattering makes only a small contribution to the total scattering in LDPE films (less than 10% of the total haze value), but has a greater effect in LLDPE films. Internal crystallisation effects will be affected by extrusion conditions in the same way as the surface crystallisation haze.

**COMBINED EFFECT OF EXTRUSION VARIABLES**

From the discussion on extrusion and crystallisation haze, it is evident that for LDPE type film, changes in some parameters, particularly freeze line distance, blow-up ratio and output rate, have opposing effects on these two components of haze. This is demonstrated in Figure 2, with extrusion haze decreasing and crystallisation haze increasing with increasing freeze line distance. The general effects of other extrusion variables are shown by the arrows in Figure 2b. Figure 5 shows the combined effects of output rate and blow-up ratio on the haze of a 38 mm LDPE film.

On the other hand, with film extrusion of LLDPE, the dominating effect of crystallisation haze results in a less complex behaviour, with worse optical properties being obtained as the freeze line distance and blow ratio are increased.

Rapid cooling (a low freeze line) assists in achieving the best optical properties with LLDPE.

Table 1 summarises the effects of varying the processing conditions on the optical properties of LDPE, LLDPE and mLLDPE films, and may be of assistance in selecting the appropriate conditions for any application.

Usually, the optical properties of HDPE rich films are not important so the extrusion conditions tend not to be altered.

![Figure 5. Variation of haze with blow-up ratio and output rate for LDPE film of MFI 4.0, density 0.925 g/cm³, film thickness 38 μm](image)
**SURFACE DEFECTS**

**Surface Melt Fracture (Sharkskin)**

“Sharkskin” is a visco-elastic surface defect caused by melt fracture at the die exit and consists of curled ridges normal to the machine direction of the film.

Low MFI narrow molecular weight distribution LLDPE is more prone to this defect which is observed at much lower output rates than with LDPE. In this context, it is commonly called surface melt fracture. The defect is an interface phenomenon and arises because of a change in the flow pattern of the polymer melt at the die wall and exit above a critical shear stress. This gives a sharkskin type surface roughness which creates poor optical appearance and may affect mechanical properties of the film. The surface melt fracture is influenced by output rate, extrusion temperature and die gap.

While the effects of surface melt fracture can be reduced by decreasing the shear rate in the die lips and increasing the melt temperature, a processing aid is often included in the film formulation to suppress the initiation of this defect and extend the maximum output rate. It is believed that the processing aid acts by coating the metal surfaces of the die, modifying the flow patterns at these surfaces.

**Orange Peel**

Orange peel (see Figure 6) is a gross surface irregularity which affects the see-through clarity of PE film. It is thought to be the same structure as sharkskin except that it is elongated in the machine direction and of coarser texture. The defect arises because of the fracture of the surface by the stresses imposed as the surface layers of the melt accelerate at the exit of the die.

The severity of the orange peel increases with an increase in output rate or a decrease in temperature. Its effects can be reduced by a higher freeze line distance, as this allows the melt a longer period to relax and the irregularities to smooth out. Greater drawdown rates and higher blow ratios also tend to smooth out this defect.

**Grain**

Grain (see Figure 7) is caused by melt inhomogeneity and is composed of discrete lumps of material of higher molecular weight than the remainder of the film projecting out of the surface. Extrusion conditions have little effect on this defect, although some improvement can be obtained by increasing the amount of homogenisation in the extruder e.g. by cooling the screw, increasing the back pressure, or improving the screw design.

### HOW THE RESIN SUPPLIER CAN HELP

Qenos offers a full range of PE resins for film packaging applications including Alkathene LDPE, Alkatuff LLDPE, Alkamax mLLDPE and Alkatane HDPE. These film grades are specifically designed for the extrusion of blown and cast film and for high performance in the subsequent conversion of the film.

Qenos PE film grades cover a wide range of Melt Flow Index and density and are specially formulated with additive packages to ensure they meet the processing, handling and end use requirements of a wide range of film applications.

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**Table 1. Effects of varying processing conditions on film optical properties**

<table>
<thead>
<tr>
<th>Processing Condition</th>
<th>LDPE</th>
<th>LLDPE and mLLDPE</th>
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</thead>
<tbody>
<tr>
<td>Higher melt temperature</td>
<td>Defects reduced, appearance improved</td>
<td>Defects reduced, appearance improved</td>
</tr>
<tr>
<td>Higher freeze line</td>
<td>Defects decrease but crystallisation haze higher</td>
<td>Crystallisation haze higher, appearance worse</td>
</tr>
<tr>
<td>Greater blow-up rate</td>
<td>Appearance improves at low freeze line and low melt temperature, but then become worse</td>
<td>Appearance worse if freeze line constant</td>
</tr>
<tr>
<td>Higher output rate</td>
<td>Defects worse but crystallisation haze reduced</td>
<td>Appearance improves</td>
</tr>
<tr>
<td>Thinner gauge (increased haul-off speed)</td>
<td>Defects decrease and appearance improves</td>
<td>Appearance improves</td>
</tr>
</tbody>
</table>

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**Figure 6. Basic types of surface irregularities in PE film – orange peel**

**Figure 7. Basic types of surface irregularities in PE film – grain**
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