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Title:
Die Geometry and Polymer Processing Additives (PPAs) Efficiency

Abstract:
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- Die Gap
- Shear Rate
- Backpressure
- Output

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Polymer Processing Additives

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Die Geometry and Polymer Processing Additives (PPAs) Efficiency

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ABSTRACT

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INTRODUCTION

The extrusion of molten polymers is often limited by the occurrence of melt fracture (MF). Although there is more than one phenomenon described by this term, sharkskin is the most common melt fracture type for film grade linear low density and high-density polyethylene (LLDPE and HDPE respectively). Sharkskin is an undesirable surface roughness on extruded plastic articles, often also referred to as orange peel or matte. It limits production rates and harms film physical and optical properties. Sharkskin is a problem that will occur upon increasing the shear rate during extrusion. It is also temperature dependent, and lowering the temperature can exacerbate the problem[1].

The extrusion condition on a processing line will be chosen to minimize this problem. One way to circumvent the problem is to increase the die gap, thus reducing the shear rate and minimizing the sharkskin. However, narrow die gaps have many advantages including: providing equipment flexibility, improved MD tear, improved gauge control, improved bubble stability and improved optics[2]. The temperature can be increased, but this often negatively affects bubble stability, or may not be an option depending on the cooling capacity.

An alternative to increasing the die gap or the temperature is to use a polymer processing additive (PPA). PPAs provide a slip layer inside the die that will alleviate sharkskin and will also reduce backpressure[3]. This allows the use of narrow die gaps, therefore reducing the need for draw down which in returns yields better physical properties[4]. An additional benefit of using a PPA is the reduction of gel formation[5] or die build-up elimination[6].

A parameter that has not previously been evaluated was the impact of the change in die gap on the formation of the slip layer inside the die. The goal of this study was to evaluate the importance of the die gap on time required for the PPA to coat the die metal. The amount of PPA required to coat the die was also evaluated.

One of the difficulties in such an analysis is the comparison point between the two die gaps may be equipment and extrusion condition dependant. As an example, one could chose to compare the results at the same output or the same shear rate. The results obtained in those two cases would be different.

Because some extruders are backpressure limited while others are output limited, we chose to do this comparison under both conditions. We also compared the two dies at constant shear rate and at constant average melt velocity.

EXPERIMENTAL

Materials

The polyethylene resin studied herein was commercially obtained. It was a well-stabilized octene copolymer of LLDPE with a 1MI and a 0.920g/cm^3 density. Only one commercially available PPA was used: Dynamar™ Polymer Processing Additive FX 9613. For the purposes of this paper. this product will be referred to as PPA-1. The PPA-1 was added using 3% masterbatch that was commercially obtained.

Equipment

Melt fracture (MF) elimination trials were conducted using a Kiefel blown film line with a 40 mm, 24/1, grooved feed extruder. The die was of spiral design with a 40 mm diameter and 0.6 mm or 0.9 mm die gap (24 or 35 mil).

Test Method

Between each test, the extruder and die surfaces were cleaned by purging the extruder with a 70% $CaCO_3$ masterbatch for half an hour. The extruder was then allowed to equilibrate with a 'barefoot' (no processing aid) control resin at the selected output and the gate pressure was monitored. At the beginning of each test, the film was fully fractured and the gate pressure was stable and consistent between tests.

For each test, the additive was added at a level of 500ppm and the percentage of the film surface that was fractured was monitored with time as a measurement of the coating of the die by the PPA-1. We will report either the time for melt fracture elimination or the coating time.

A series of outputs were selected to allow a direct comparison between the two dies, while keeping either the shear rate, the output, the backpressure, or the melt velocity constant. Table I summarizes the conditions tested.

RESULTS AND DISCUSSION

Melt Fracture Elimination

As a forewarning, it should be pointed out that this study was done in a well-stabilized LLDPE with no other additives than the antioxidant package. For instance, the presence of antiblock, slip, or light stabilizer is likely to yield different results [7,8,9]. Similarly, synergists such as antioxidants may show a different correlation[10].

It must also be pointed out that we are describing here the coating process of the die. The correlation between coating time and maintenance level of PPA-1 is not described here and will hopefully be part of a future study.

The melt fracture elimination curves for the six conditions given in Table I are shown in Figure 1. The results given here clearly show that the coating process is not independent of the process conditions.

As an example, with the 0.9 mm die, the rate of melt fracture elimination increases with the output This is also observed with the 0.6 mm die, however, there is only a small difference between the 6.8kg/h (B) and 13.6/kg/h (C) data. It can also be noted when comparing the two dies that the time to eliminate melt fracture is not solely dependent on output

Each pair of data can also be compared. Test C and Test F are both at 13.6kg/h. However, the coating time is shorter on the 0.6 mm die. This is an indication that the coating process is not a simple matter of mass throughput. We will expand more on this, later in this article.

When comparing Test B and Test E, or Test A and Test D, (Tests that exhibit the same melt velocity), again, the narrower die provides a more efficient coating process. A similar trend is obtained for constant backpressure when comparing Test B and Test D.

When comparing Test B and Test F, the overlap between the two curves is fairly good. This would indicate that the shear rate is the main factor controlling the coating process.

To confirm that the coating time is not directly related to throughput, we normalized the coating curves and plotted them as a function of the amount of PPA-1 going through the die on Figure 2. Again, if the coating time was solely output controlled, all the curves would overlap.

In Figure 2, all the Tests with the 0.6 mm die require less PPA-1 than the Tests with the 0.9 mm die. This is an indication that only the portion of the PPA-1 that is close to the die wall is used in the coating process. The PPA-1 in the core of the extrudate does not contribute to the melt fracture elimination. In the narrower die, there is less core material; therefore, the PPA-1 is used more efficiently.

We must point out that previous results[10] indicated that for constant shear rate, increasing level of PPA-1 could be normalized to a constant level. This indicates that the amount of PPA-1 going through the die is important but the conditions of extrusion are playing a role as well.

Another point to notice on Figure 2 is that for the 0.9 mm die the amount of PPA-1 required is proportional to the throughput. However, this is not true for the 0.6 mm die where the Test B (6.8 kg/h) requires the least PPA-1. We suspect that the Test C (13.6 kg/h) corresponds to a shear rate (639/s) that is too close to the onset of cyclic melt fracture (CMF). This disturbs the coating process.

Gaussian Analysis

In order to get a better understanding of the coating process, we tried to analyze the coating process as a statistical phenomenon. In that case, the droplets of PPA-1 would reach the die in a random fashion (for a given set of conditions), and the die coverage would follow a normal distribution.

If this assumption were valid, a probability plot would lead to a linear relation. The data from Figure 1 was plotted using a probability scale on Figure 3. From the regression, one can obtain the average time (time to 50% MF, $t_{1/2}$) and the standard deviation (sigma). The results for those two regression parameters are reported in Table II. For this study, we selected $t_{1/2}$ as a comparative factor for the coating time. $t_{1/2}$ is a statistical average of the whole coating curve and should be a good representation of the coating process.

From these regression parameters, one can calculate the statistical coating curve. This is shown on Figure 4, where the agreement between the calculated curves and the raw data is good. Results from Figure 3 are indeed following a linear relation consistent with a Gaussian process. Similarly, Figure 4 indicates that the Gaussian analysis is a good first approximation for the process we are describing here.

When plotting the time to 50% MF vs. the output, the results obtained from each die are well differentiated on Figure 5. This is again indicative of the coating process being correlated with the shear rate rather than the output, as shown on Figure 6.

It must be pointed out that for a single die, within the linear region of Figure 6, one cannot differentiate between shear rate and output. It is only when comparing two dies, that one can see the importance of the shear rate.

CONCLUSIONS

We evaluated the effect of the die gap on the coating process of a PPA on a film-blowing die. The effect of varying the shear rate, the backpressure, the melt velocity, and the output, was monitored for two die gaps. The time to eliminate melt fracture and the amount of PPA-1 required to do so showed no correlation with the melt velocity or the backpressure.

However, there is some correlation between the time to eliminate melt fracture and the output. This correlation exists only below a maximum shear rate (below 350/s in our data set). Furthermore, this correlation is valid for a single die only. When comparing data from two dies, the shear rate is a better descriptor of the coating process. The correlation is then independent of the die selected.

The data was also analyzed using a Gaussian approach. The correlation with the shear rate was again clearly demonstrated when plotting the time to eliminate 50% of the melt fracture. In this case, we obtained a linear correlation.

However, at high shear rates, the data does not follow the same trend. We suspect that to be linked to the onset of CMF. At those high shear rates, the higher stresses and the perturbations from the CMF would disturb the coating process and increase the coating time.

The throughput was shown in the past to be one of the main factors in reducing the coating time (constant conditions, increasing levels)[10]. The results obtained here indicate the shear rate also has a significant effect. At higher shear rates, the PPA-1 is coating the die more efficiently. This phenomenon is probably linked to the flow profile in the die and related to the stress gradient across the die.

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Table I: Processing Conditions

Test Conditions	A	B	C	D	E	F
Gap (mm)	0.6	0.6	0.6	0.9	0.9	0.9
Output (kg/h)	5.4	6.8	13.6	8.2	10.0	13.6
Shear Rate (1/s)	256	320	639	180	220	300
Av. Velocity (cm/s)	2.59	3.24	6.48	2.67	3.26	4.44
Backpressure (MPa)	6.34	7.79	12.2	7.79	9.17	11.1

Table II: Regression Parameters from the Gaussian Analysis

Conditions	$t_{1/2}$ (min)	Sigma (min)
A: 5.4kg/h, 0.6 mm	83.5	64.1
B: 6.8kg/h, 0.6 mm	41.5	30.9
C: 13.6kg/h, 0.6 mm	33.4	23.7
D: 8.2kg/h, 0.9 mm	215.7	144.9
E: 10.0kg/h, 0.9 mm	125.6	73.3
F: 13.6kg/h, 0.9 mm	51.5	27.9

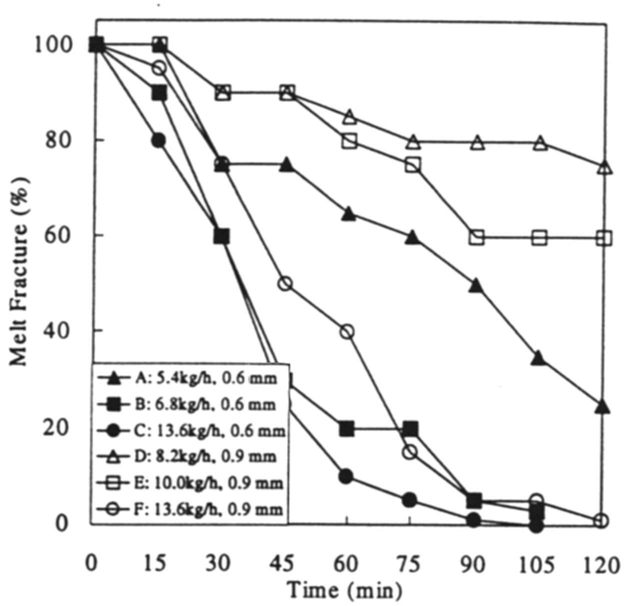

Figure 1: Melt Fracture Elimination vs. Time

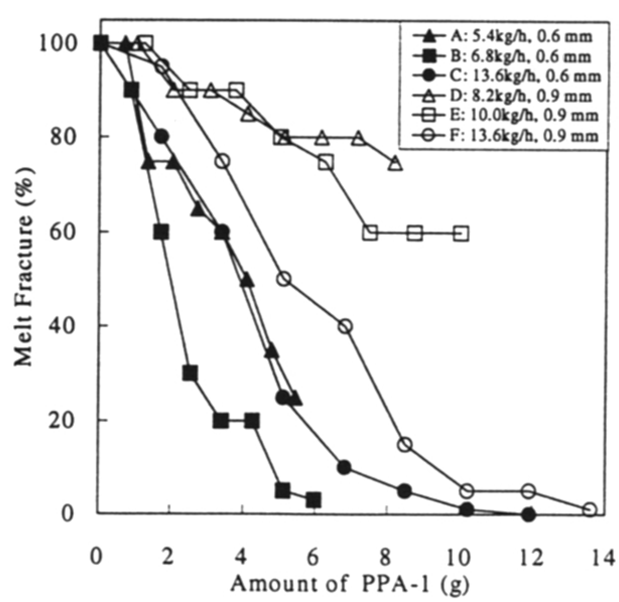

Figure 2: Melt Fracture Elimination vs. Mass of PPA-1

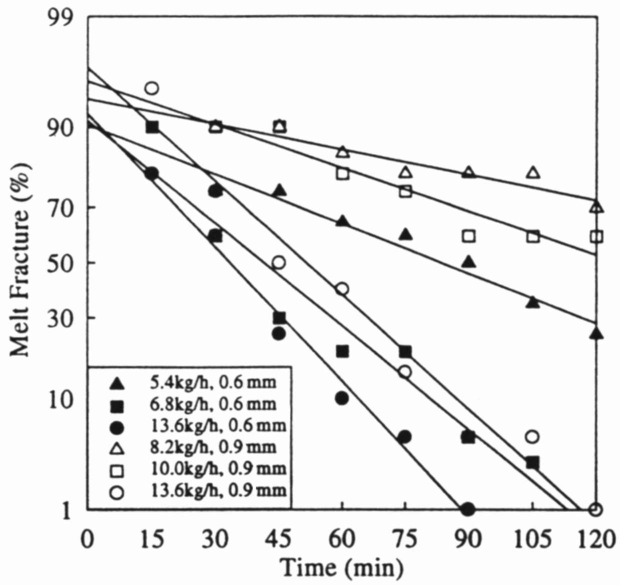

Figure 3: Probability Plot for Melt Fracture Elimination vs. Time

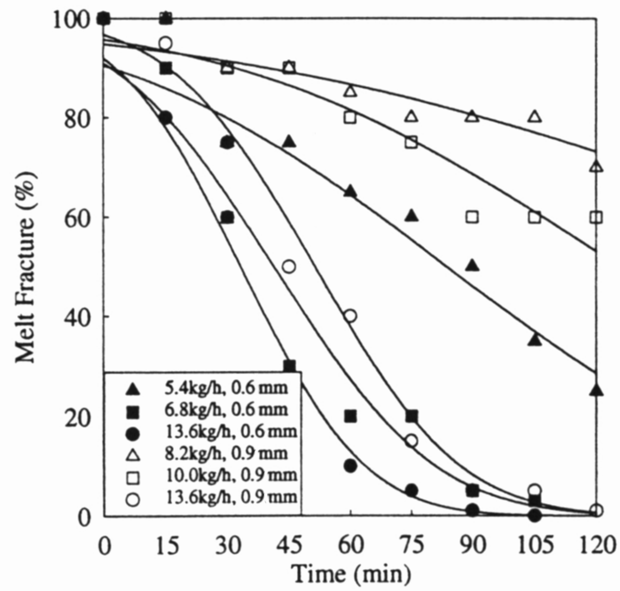

Figure 4: Calculated Melt Fracture Elimination Curves vs. Time

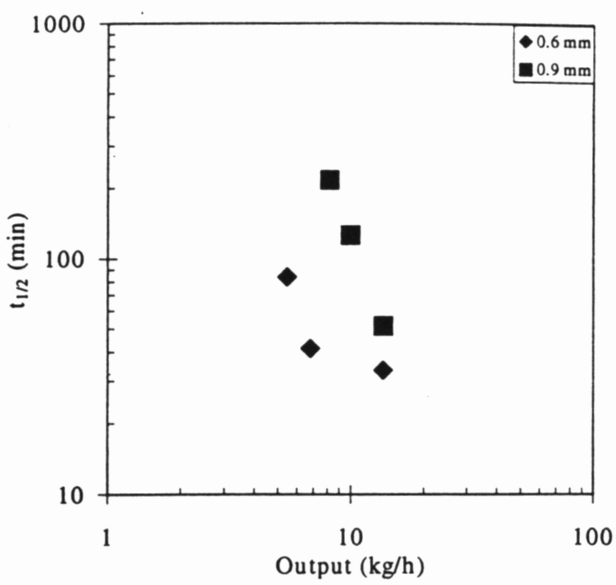

Figure 5: Time to Reach 50% MF vs. Output

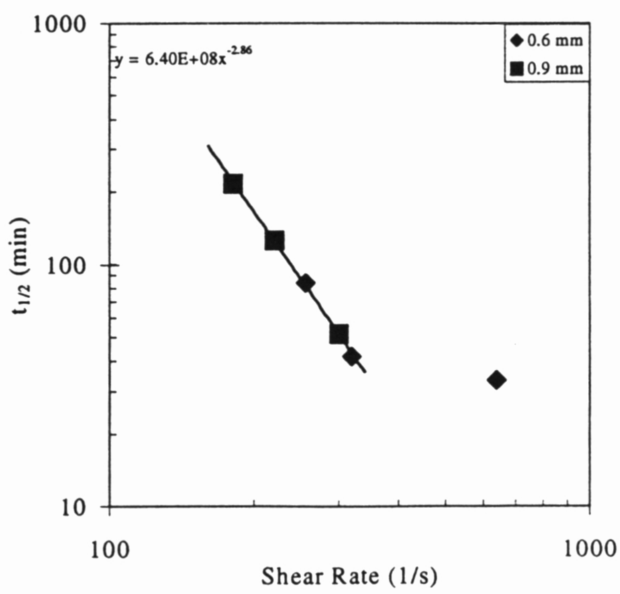

Figure 6: Time to Reach 50% MF vs. Shear Rate

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