DIE LIP BUILD-UP IN THE FILLED LOW DENSITY POLYETHYLENE WIRE AND CABLE EXTRUSION

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ABSTRACT

It is common that a highly filled master batch (MB) contains a low molecular weight (LMw) lubricant as a filler dispersion agent and compounding processing aid. Sometimes, the presence of such a lubricant in the MB creates a die build-up problem in the extrusion process. Several MBs with a laboratory Banbury mixer were produced to investigate the effect of the amount of aluminum stearate (as a lubricant) on dispersion and die build-up. When the MB contains the stearate below the critical amount, die build-up decreases and filler dispersion improves with increased amounts of stearate. At the critical amount of the stearate, the MB shows good dispersion as well as minimum build-up. Above the critical amount, dispersion is still good. But, build-up increases with further increase in the amount of stearate added up to 3.0%. This observation results from the different mechanisms of the die build-up, which depends on whether the stearate level is below or above the critical amount. We confirmed such a mechanism in the commercial extrusion process of the MBs. This observation leads to the conclusion that in the use of the stearate (as a dispersion agent) for highly filled MB, the amount of the stearate need to be optimized to minimize a die build-up of the MB with good filler dispersion.

■ **KEYWORDS**
Die Build-up, Due Lifetime, Stearate, Master Batch, LDPE, Dispersion Agent, Stability, Filler Dispersion

INTRODUCTION

Die lip build-up, also called plate-out, die drool, die drip, or die peel, refers to the unwanted accumulation of material on the open face of wire and cable extrusion dies. Build-up also creates die streaking onto a surface of an extruded product.

Excessive die lip build-up has been an issue in the wire and cable as well as the plastic industry [1-4]. This is due to the fact that die build-up diminishes both the aesthetic quality and the properties of a product. For an example, a reattached build-up of material in a wire coating extrusion may diminish the dielectric properties of the insulating material [1]. A shorter die lifetime resulting from a quicker die lip build-up requires more frequent downtime. If an extrusion operator observes the die build-up during extrusion, the operation must be stopped for die cleaning. Thus, such an extrusion is not desirable and costly.

Low molecular weight (LMw) materials such as polyolefin wax, silicon oil, or metal stearate have been widely used in the compounding industry to improve filler dispersion as well as the compounding operation [4-5]. Good filler dispersion is critical in the use of a MB for industrial applications. In general, the LMw material used for MB products is incompatible with the base resin of the MB. During extrusion, LMw material can preferentially migrate to the surface of the die. This behavior of LMw material migration appears to be similar to that of fluoroelastomer processing aids. However, the nature of a lubricant layer between LMw and fluoroelastomer is different, leading to the formation of die build-up for the former and elimination of build-up for the latter [6-8].

Several MBs of 50 wt% filler loading were produced using a laboratory Banbury. The laboratory extrusion test has been carried out for the measurement of the die build-up on the MBs. The objective of this work is to investigate the effect of the amount of stearate on the rate of die build-up. The build-up mechanism is proposed as a function of the amount of stearate added. The build-up data of lab scale MBs are in line with those of commercial extrusion with the MBs. The result provides a guideline to optimize the level of lubricant in the production of a MB.

EXPERIMENTAL

SAMPLE PREPARATION
A Farrel ODC Banbury mixer (2400 CC) was used to produce 50% filled MBs with the same components of filler, base LDPE resin, and antioxidants (700 PPM for the primary and 500 PPM for the secondary) but different levels of aluminum stearate.   The Banbury drop was pelletized through a 50.8 mm single screw extruder to produce finished pellets products for evaluations.

CHARACTERIZATION

■   Rheological Measurements
The dynamic rheological measurements were carried out on a Rheometrics RDA-II equipped with parallel plates. The measurements were done at 210°C with a frequency sweep from 398 to 0.0640 rad/sec with 5% of strain. We also used the plots of G′ (elastic modulus) vs. G″ (loss modulus) for a comparison purpose in the change of melt elasticity.

We obtained the apparent shear viscosity as a function of apparent shear rate for the MBs using a Kayeness capillary rheometer at 210°C.

■   Filler Dispersion Test
We used a pressure rise test (PRT) for the filler dispersion of the MBs used in this investigation. The PRT is analogous to the screen pack-plugging test, which is used to determine the quality of MBs. The test uses a Haake 90 single screw extruder (L/D=23, D=20 mm). The heated die has a breaker plate followed by the following screen pack arrangement in mesh: 60-60-325-500-60. The extruder screw was run at 150 RPM. The MB was let down to a final 12.5 wt% filler content using the let down resin (LDPE, 7 MI, 0.920 density). The pressure rise (PR) in psi is obtained by subtracting the first 5 minutes reading from the 35 minutes reading: the higher PR means poorer filler dispersion for the MB evaluated due to quicker screen pack plugging.

■   Stability Test
A thermo-mechanical stability test was carried out using a Haake Rheomix 90 at 60 RPM and 230°C for the MBs. We monitored torque variation as a function of time. The variation represents the extent of thermo-mechanical stability. We also examined the test samples for the rheological change to identify the root cause of the variation.

■   Die Build-Up Test
A Haake 25 mm single screw extruder at 125 RPM attached with 50 mm slit die with 0.5 mm die opening was used for the laboratory measurements of die build-up. A sheet extrudate was drawn with a take-up device to a 1.5:1draw ratio to minimize a die swell effect. We monitored the percent coverage of the die deposit every 5 minutes during extrusion of the MBs. Average percent coverage was taken from three measurements for each sample.

■   Infrared (IR) Measurements
The die deposit was carefully and quickly collected from the lip of the die as soon as enough build-up was noticed during extrusion. The IR spectra were acquired using a Nicolet 60SX FTIR spectrometer under nitrogen purge. The carbonyl (C=O) stretching band at 1860 – 1610 cm^{-1} was used to measure the extent of degradation. The thickness band at 2107-1976 cm^{-1} was also used for the correction of sample thickness for the spectra.

RESULTS AND DISCUSSION

Table 1 shows the dispersion and rheological data of the experimental MBs containing different amounts of the stearate. As expected, filler dispersion improves with addition of stearate up to 0.5 wt% and remains unchanged with further addition of the stearate up to 1.0%. Further addition of stearate up to 3.0% deteriorates dispersion. The dynamic rheological data indicates little difference in the frequency viscosity at 25 rad/sec for the MBs, A-1, A-2, & A-3 (Table 1) which contain the stearate less that 0.6%. However, we observed a decreased viscosity with further increased level of the stearate, as going from 0.6 to 3.0 %. We confirmed such decreased viscosities in the capillary shear viscosity data (figure 1). A decreased viscosity with further increased amount of the stearate results from a lubricating effect under shear deformation.

Table 1.   Basic, dispersion, and rheological data of the experimental MBs with different levels of the stearate.

MB	Level of Stearate (%)	PR (psi)	Complex Viscosity Poise
A-1	0.0	330	5650
A-2	0.3	207	5580
A-3	0.5	110	5520
A-4	0.6	125	5400
A-5	1.0	115	4980
A-6	1.5	140	4440
A-7	3.0	162	4010

Figure 1.   Rheological data for MB, A-3 (0.5%), A-4 (0.6%), A-5 (1.0%), A-6 (1.5%), and A-7 (3.0%).
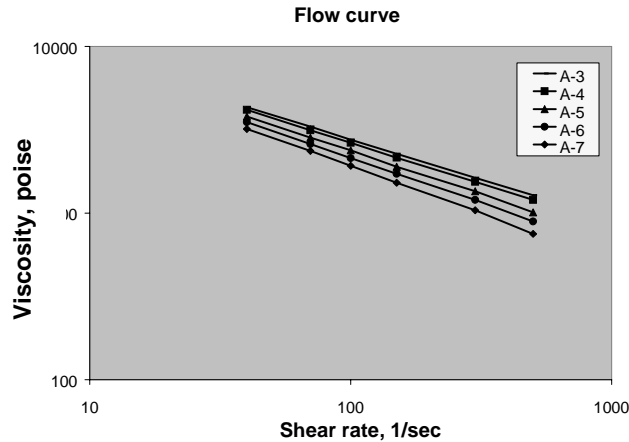

Figure 2 shows the stability data of experimental MBs. Interestingly, MB A-1 tends to show higher torque build-up with increased mixing time than A-3 during the stability test. The only difference between these two MBs is the amount of stearate. In order to identify the origin of higher torque for A-1, compared to A-3, we examined the rheology of the stability tested samples. As shown in figure 3, the tested A-1 shows much higher G' (elastic modulus) for a given G''(loss modulus) than A-3. Both MB showed very similar G' at a given G'' before the test. The higher elasticity for MB A-1, compared to A-3, can be attributed to a presence of chain extension reaction associated with degradation process. MB A-2 showed similar behavior to A-1 (figure 3) whereas MB A-4 through A–7, compared to A-3, shows much less torque with similar elasticity. The reduced torque results from more lubrication effect with higher amount of the stearate addition and subsequently, leads to the reduced degradation under the similar thermo-mechanical treatment.

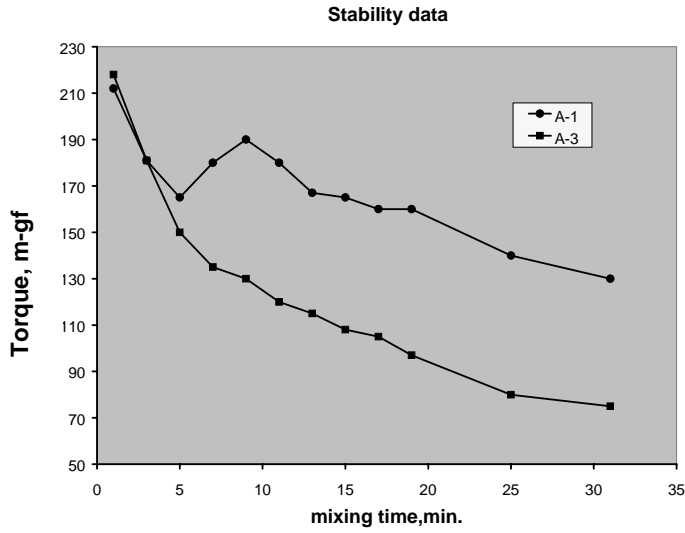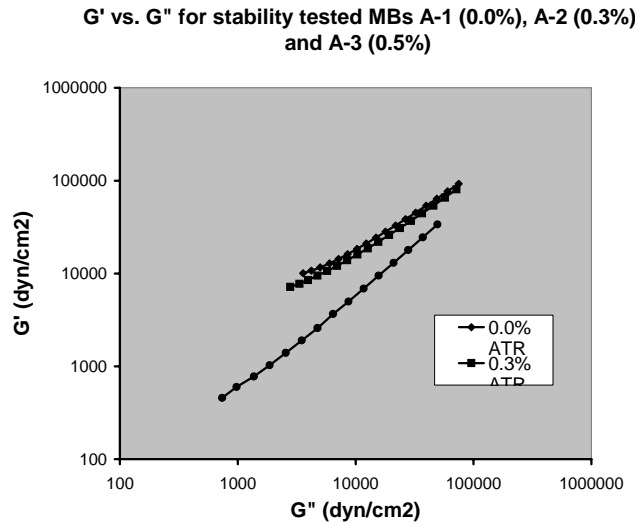**Figure 2.   Stability data for MB A-1 (0.0%) and A-3(0.5%)**

Figure 3.   Dynamic rheological data (G' vs. G") for the stability tested MB A-1 (0.0%), A-2 (0.3%) and A-3 (0.5%)

The die build-up test with the lab scale extrusion indicates that the rate of build-up decreases with increased amount of the stearate up to 0.5 % and then increases with further increased amount of the stearate (figure 4).

Figure 4.   Rate of die build-up as a function of the amounts of stearate.
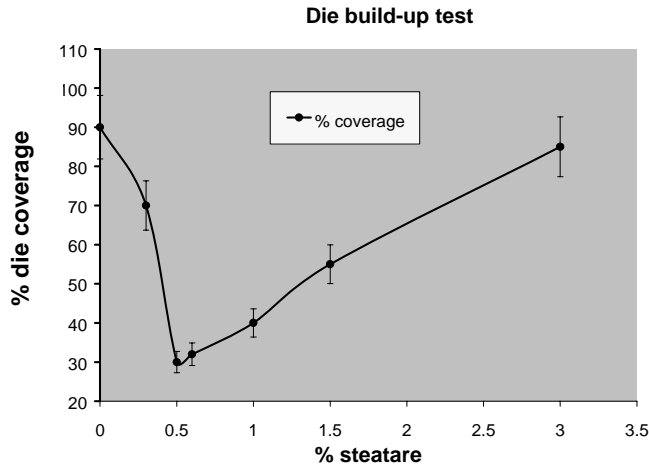
Die build-up test

This result indicates a necessity of optimized amounts of the stearate addition to minimize die build-up with a good dispersion. Coupled with the result of the stability test, the mechanism for the build-up appears to be different, depending on the amount of the stearate. The mechanism will be discussed in a later section.

Figure 5 shows the capillary shear viscosity data of the stability tested MBs A-1 to A-3. A-1 shows lower shear viscosity than A-3. Previously, it has been shown that both MBs showed very similar viscosity data before the test. The lower viscosity for A-1 is indicative of the increased amount of LMw material which was developed by the degradation process during the stability test. It has been indicated that the degradation process develops both melt elasticity and LMw material due to the presence of both reactions; chain extension and chain scission [9]. A similar analogy can be applied to the MB A-1 for more build-up of A-1 with the single screw extrusion process when compared to A-3

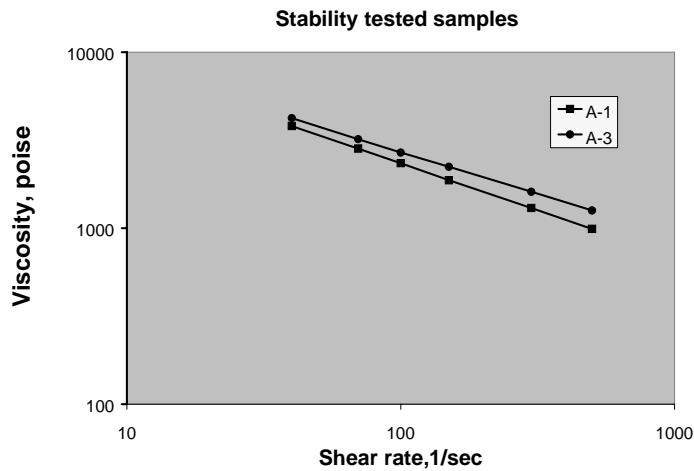**Figure 5.   Rheological data for the treated A-1 (0.0%) and A-3 (0.5%)**

Stability tested samples

Figure 6 shows the IR results of the material collected from the die build-up of the MBs. The spectra contain the carbonyl peak at 1725 cm^{-1} along with other major polyethylene peaks. The carbonyl band represents the presence of degraded product in PE [9]. Going from MB A-3 to A-1, we observed an increase in the carbonyl peak height with decreased amounts of the stearate with extrusion. This indicates that A-1 experienced more degradation present during the Haake extrusion than A-3. From A-4 to A-7, it is also shown that the carbonyl peak height increases with further addition of the stearate in the MBs. An increased carbonyl peak height with higher amounts of stearate present in the MBs (A-4 to A-7) originates not from degradation (as discussed previously) but from more stearate in the build-up. For example, more stearate comes along with extrudate for A-7 but not for A-4. The normalized data of the peak with respect to thickness confirms the observation described above (Table 2) where MB C-1 is a commercial product (see Table 3).

Figure 6.   IR data for the die deposits obtained from MB A-1 (0.0%), A-3 (0.5%), A-4 (0.6%), A-7 (3.0%), C-1 (0.3%).
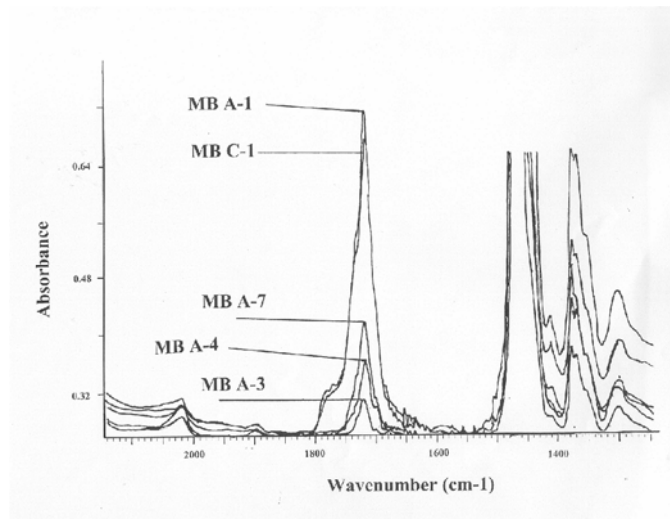
Table 2. Normalized carbonyl band with respect to the IR samples of the die deposit collected from extruding the MBs.

MB	% Stearate	Carbonyl Band Height	Height of Thickness Band	Normalized Ratio
A-1	0.0	30.9	1.4	21.3
A-3	0.5	1.0	1.0	1.0
A-5	1.0	2.2	0.7	3.2
A-7	3.0	4.1	0.7	5.7
C-1	0.3	20.5	1.4	14.8

Table 3 exhibits the basic data and die build-up performance of commercial MBs, C-1 and C-2. Both MBs were produced with identical compositions and compounding conditions but different levels of the stearate: 0.3% for C-1 and 0.5% for C-2. The MBs were letdown to 12.5% filler content for the extrusion evaluation. Both MBs show very similar filler dispersions but much difference in die lifetime. This observation of the die lifetime from the commercial extrusion for the MBs is in line with that of the lab scale build-up measurements (figure 4).

Table 3. Basic, dispersion, and die lifetime data of the commercial MBs.

MB	% Stearate Level	PR, psi	Die Lifetime, hrs
C-1	0.3	178	2
C-2	0.5	108	12 +

PROPOSED MECHANISM FOR THE DIE BUILD-UP

Based on the results thus far, the critical amounts of stearate were 0.5 wt% for the MB with a minimized build-up and good filler dispersion. It was also found that at amounts of stearate below 0.5%, degradation process promotes build-up. Above that amount, the excess stearate appears to promote the build-up.

Figure 7 describes a proposed model for the origin of die build-up depending on the amount of the stearate. At the amount below 0.5%(figure 7(a)), since there is not enough stearate to cover all filler surfaces, there may be a lot of friction in the interface between the filler and the matrix during mixing/extrusion, leading to a higher extent of degradation. Such degradation promotes both reactions; chain extension and chain scission, leading to enhanced melt elasticity and creating the LMw material. The newly developed LMw is responsible for the build-up during extrusion. At higher amounts of the stearate (>> 0.6%)(figure 7(b)), we can expect good incorporation of the stearate with filler surface which minimizes friction due to the lubricant effects. However, the amount of stearate, which does not incorporate with filler surface, remains in the matrix as excess. Since the amount of filler used for the MBs was fixed, an increased amount of the stearate addition for finished MB represents an increased amount of excess stearate. Such an excess amount of the stearate leads to the formation of die build-up during extrusion.

Figure 7.   Proposed model for the mechanism o: (a) stearate << critical amount (b) stearate >> critical amount.
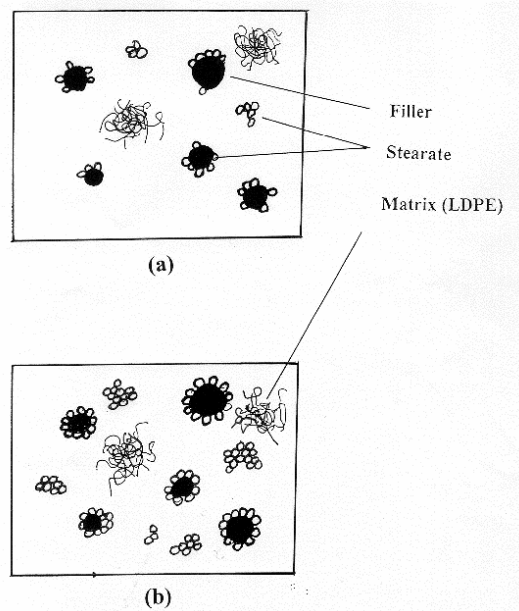
CONCLUSIONS

In the use of the highly filled master batches for extrusion applications, the filler dispersion and extent of die build-up depend on the amount of the stearate. We identified the critical amount of the stearate showing good dispersion as well as minimized die build-up. The critical amount was 0.5 wt% for the MBs investigated for this work. At an amount less than 0.5 wt%, thermo-mechanical degradation promotes die build-up; the build-up increases with decreasing amount of the stearate. The increased die build-up relates to the increased presence of the LMw material developed by the chain scission with the degradation process. At the higher amounts of stearate addition (above 0.6%), the large amounts of stearate create higher rates of die build-up even though the stability increases due to more lubrication: the build-up increases with increased amounts of stearate. This result is in line with the observations from commercial extrusion.

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