On Formation Mechanism of Die Drool and Slip-stick Phenomena during Linear HDPE Polymer Melt Extrusion

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Abstract: In this work, die drool and slip-stick flow phenomena are experimentally investigated for two linear high density polyethylene melts differing in the high molecular weight content only. It has been found that slight increase in the high molecular weight fraction can significantly promote the onset and intensity of both investigated flow phenomena.

Key-Words: Die drool, Slip-stick, Flow induced molecular weight fractionation, Flow instabilities, Elasticity, Extrusion, Polymer melt.

1 Introduction
Extrusion is one of the most commonly used polymer processing technique for creating of continual products such as pipes, profiles, films, cables etc. During this process, polymer melt prepared from solid pellets inside the extruder is pushed through the extrusion die in which the final product is formed. Inside the extrusion die or at the die exit edge vicinity, different types of flow instabilities can take place \cite{1, 2}. Die drool and slip-stick phenomena are some of them.

The first, die drool (also called die lip buildup, drooling, die bleed, die plateout, die deposit, die drip or die moustache) is in extrusion art defined as unwanted spontaneous accumulation of extruded polymer melt at die exit faces. Such accumulated material is generally named as drool. Within extrusion time, this drool builds up into a large mass, frequently breaks away from the die, completely (or partly) encloses extruded product, adheres perseveringly onto extruded product surface and thus damage it. If this phenomenon takes place during the extrusion process for particular polymer system, it is necessary to periodically switch off the extrusion line in order to complete die cleaning which is uneconomical and also time consuming. A lot of research papers focused on this unwanted extrusion phenomenon have been already published \cite{3-11} and several factors have been found to support its appearance. They include pressure fluctuations in screw \cite{3}, volatilities, low molecular fractions of the polymer, fillers, poor dispersion of pigments \cite{4}, die swell \cite{3, 5}, processing near degradation temperature \cite{5, 6}, dissimilar viscosities in blends \cite{7}, improper die design and die material \cite{8, 9}, negative pressure at the die exit \cite{8}, utilization of linear rather than branched polymers \cite{10}, high polymer melt elasticity \cite{11}, etc.

The second phenomenon, slip-stick, appears at very narrow processing window (depending on temperature and mass flow rate for given polymer melt) in which the wall shear stress/pressure in the extrusion die oscillate between two extreme values, although the imposed mass flow rate is kept constant. Number of research works focused on this instability have already been published \cite{12-14} and many hypothesis aiming to explain their source and formation mechanism have been formulated \cite{1}.

Recently, it has been found that die drool phenomenon during the HDPE melt extrusion at low temperatures starts to appear at the slip-stick instability region \cite{10, 11} suggesting that both flow instabilities can be caused by the flow induced molecular weight fractionation inside the extrusion die occurring at sufficiently high shear stresses during which polymer macromolecules are redistributed during the flow according to their lengths (short chains near the die wall, long chains in the middle of the flow channel).
In this work, the above describe hypothesis is tested for die drool and slip-stick flow phenomena utilizing two carefully prepared virtually identical linear HDPE polymers differing in very long linear molecules amount only.

2 Experimental

In this work, two stabilized unfilled virtually linear high density polyethylene polymer melts having broad molecular weight distribution were chosen (HDPE Liten FB 29 E2009 3220 4479 signed thereinafter in the text as HDPE 1 and HDPE Liten FB 29 E2009 5498 7937 signed as HDPE 2). Both polymers are extrusion grades from Unipetrol RPA company, Czech Republic with basic characteristics summarized in Table 1.

Table 1. Basic characterization of chosen HDPEs.

<table>
<thead>
<tr>
<th>Property</th>
<th>HDPE 1</th>
<th>HDPE 2</th>
</tr>
</thead>
<tbody>
<tr>
<td>Density (g.cm(^{-3}))</td>
<td>0.950</td>
<td>0.950</td>
</tr>
<tr>
<td>(M_n) (g.mol(^{-1}))</td>
<td>17,045</td>
<td>16,115</td>
</tr>
<tr>
<td>(M_w) (g.mol(^{-1}))</td>
<td>239,300</td>
<td>228,350</td>
</tr>
<tr>
<td>(M_z) (g.mol(^{-1}))</td>
<td>1,613,000</td>
<td>1,412,000</td>
</tr>
<tr>
<td>(M_{z+1}) (g.mol(^{-1}))</td>
<td>3,476,000</td>
<td>2,826,000</td>
</tr>
<tr>
<td>(M_w/M_n) (-)</td>
<td>14.05</td>
<td>13.94</td>
</tr>
<tr>
<td>Melt flow index (g/10min)</td>
<td>0.15</td>
<td>0.15</td>
</tr>
<tr>
<td>Zero-shear rate viscosity (Pa.s)</td>
<td>670,000</td>
<td>380,000</td>
</tr>
<tr>
<td>Flow activation energy (kJ.mol(^{-1}))</td>
<td>13.817</td>
<td>14.031</td>
</tr>
<tr>
<td>DSC peak melting point (°C)</td>
<td>130.8</td>
<td>130.7</td>
</tr>
<tr>
<td>DSC heat of fusion (J.g(^{-1}))</td>
<td>234.9</td>
<td>242.6</td>
</tr>
<tr>
<td>Crystallinity (%)</td>
<td>80.16</td>
<td>82.81</td>
</tr>
</tbody>
</table>

Taken from the original material data sheet for basic polymer HDPE Liten FB 29.

2.1 Rheological Characterization

In this work, stable-state shear and uniaxial extensional viscosities and first normal stress coefficient (representing the level of melt elasticity) were determined for both polymer melts. In more detail, shear viscosity at low shear rates was measured on rotational rheometer Advanced Rheometric Expansion System ARES 2000 (Rheometrics Scientific, USA) in parallel plates configuration and for high shear rates region twin-bore capillary rheometer Rosand RH7-2 (Rosand Precision, United Kingdom) in circular capillary mode was used. Further, uniaxial extensional viscosity was determined by Sentmanat Extensional Rheometer SER-HV-A01 model (Xpansion Instruments, USA [15]) attached to ARES 2000. Finally, first normal stress coefficient was determined on capillary rheometer Rosand RH7-2 equipped by slit die with utilizing Han’s exit pressure drop methodology [16]. It should be noted that only data above wall shear stress of 25kPa [17] at which the pressure transducers error is not significant were used.

All rheological properties were determined at one reference temperature 190°C. The results are shown in Figs. 1 and 2.

![Fig. 1. Stable-state uniaxial extensional and shear viscosities for both HDPEs determined at 190°C.](image)

![Fig. 2. Stable-state normal stress coefficients for both HDPEs measured at 190°C.](image)
higher uniaxial extensional/shear viscosities and also
elasticity (first normal stress coefficient) than HDPE 2.

2.2 Die Drool Measurements

For die drool evaluation of both polymer melts,
specially designed laboratory extrusion line was
used (see Fig. 3). From right to left, the line is consisted of Plasti-Corder 2000 (Brabender,
Germany) power unit, single screw extruder with
diameter \( D = 30 \text{ mm} \) and \( L = 25D \) (standard
single-thread screw with compression ratio 4:1, and
lengths of zones: feed \( L_1 = 10D \), compression
\( L_2 = 3D \), metering \( L_3 = 12D \)), specially designed
extrusion die with circular replaceable capillary
(firstly introduced in [18]), tripod with photo camera
in order to visualization die drool, draw off
mechanism and finally data processing unit.

The extruder heating zones (from the hopper to
the die) were heated to \( T_1 = 150°C, T_2 = 155°C, T_3 = 160°C \) and \( T_4 = 160°C \), respectively by keeping
the annular tube (connecting die and extruder) and
die exit temperature constant, \( T_5 = T_6 = 160°C \).
It should be mentioned that this temperature profile
was chosen with respect to DSC calorimetry curves
to ensure both materials are totally melted and
simultaneously, high level of elastic energy
promoting die drool is stored in both melts [10, 19].

To investigate relationship between die drool and
slip-stick phenomena, mass flow rates (MFRs) for
both materials were varied between 0.075 kg.hr\(^{-1}\) up
to 0.725 kg.hr\(^{-1}\) \( (\text{i.e. from } 68 \text{ s}^{-1} \text{ up to } 652 \text{ s}^{-1} \text{ in term of apparent shear rates}) \). Drool mass evaluation
method was based on direct collection of drool from
die exit face by a tweezer and then its weighting on a
sensitive analytic balance which we repeated 3 times
to ensure a reproducibility of such data. Finally,
extrusion time was chosen 10 minutes for each test.

2.3 Slip-stick Measurements

For precise wall shear stress/pressure oscillations
evaluation capillary rheometer Rosand RH7-2
(Rosand Precision, United Kingdom) with capillary
diameter of 0.5 mm and \( L/D \) ratio of 16 was used.
Apparent shear rates during tests decreased from
600 s\(^{-1}\) down to 50 s\(^{-1}\) (20 stages logarithmically
scaled). Testing temperature was 160°C (same as die
temperature during die drool tests).
3 Results and Discussion

As the first, we performed die drool tests. The drool mass were recalculated from weight to a dimensionless form of buildup ratio \((BR)\), firstly introduced by Gander and Giacomin [20]:

\[
BR = \frac{\dot{B}}{m}
\]

(1)

where \(m\) is total mass flow rate of extruded polymer melt and \(\dot{B}\) means buildup rate:

\[
\dot{B} = \frac{B}{t_e}
\]

(2)

where \(B\) is the mass of accumulated die drool material on the die exit face and \(t_e\) is total extrusion time of each test (10 minutes in our case).

Speed of extrusion we express as apparent shear rate:

\[
\dot{\gamma}_{\text{app.}} = \frac{4Q}{\pi R^3}
\]

(3)

where \(Q\) represents volumetric flow rate and \(R\) means radius of used capillary (in our case 0.8 mm).

The results of die drool tests are shown in Fig. 4. It is clearly seen that HDPE 1 (the sample containing high molecular weight fraction) produces significantly higher drool mass than HDPE 2 (the sample with small amount of high molecular weight fraction).

![Fig. 4. Dimensionless buildup ratio for both HDPEs as function of apparent shear rate.](image)

Die drool visualization at the die exit region for the increased apparent shear rate for both tested HDPE samples is provided in Fig. 5.

![Fig. 5. Die drool visualization for HDPE 1 (left hand side column) and HDPE 2 (right hand side column) for first five apparent shear rates (extrusion direction is from right to left).](image)

The slip-stick intensity for both samples was quantified through an apparent wall shear stress difference (calculated from oscillating raw pressure data) according to the following equation:

\[
\Delta \tau_W(\dot{\gamma}) = \tau_{W,\text{MAX}}(\dot{\gamma}) - \tau_{W,\text{MIN}}(\dot{\gamma})
\]

(4)

where \(\tau_{W,\text{MAX}}(\dot{\gamma})\) and \(\tau_{W,\text{MIN}}(\dot{\gamma})\) represent the maximum and minimum wall shear stresses, respectively, at the given apparent shear rate at which the pressure oscillations take place. Just note that the wall shear stress difference is equal to zero for apparent shear rates at which pressure/wall shear stress is not oscillating for particular apparent shear rates.
Fig. 6. Wall shear stress difference as a function of apparent shear rate for both materials.

This variable is plotted as the function of apparent shear rate for both samples in Fig. 6. Clearly, the wall shear stress difference for HDPE 1 is higher than for the HDPE 2 as well as the onset of non-zero wall shear stress difference is shifted for HDPE 1 towards the lower apparent shear rates in comparison with sample HDPE 2.

The obtained results indicate that slight increase in the high molecular weight fraction of HDPE 1 polymer (leading to melt elasticity increase) leads to “more effective” short chains movement toward the wall in comparison with the other polymer HDPE 2 which is in good agreement with our previous work [10] and also original Busse’s theory about flow induced molecular weight fractionation [19].

4 Conclusion

In this experimental work, flow stability of two linear HDPE polymer melts differing in high molecular weight fraction content was investigated. It has been revealed that slight increase in HDPE high molecular weight fraction (which increases melt elasticity) may significantly promotes onset and intensity of both, die drool as well as slip-stick phenomena.

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References:


