About the origin of sharkskin

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With 11 figures and 3 tables

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1. Introduction

Flow instability occurs in polymer melts passed through a die when the shear stress at the wall reaches a critical value \( S^* \) (1). Since it has been discussed for about 30 years and ample reviews have recently been published (1, 2), we do not present the whole subject here, but confine our attention to the main flow instability occurring in HDPE (linear polyethylene) in a capillary, and to the so-called “sharkskin” effect, which is a small-scale high frequency distortion of the extrudate.

Bagley et al. (3) observed that in HDPE flowing through tubes the output value became a double-valued function of the extrusion pressure over a limited range (see fig. 1). Due to that particular relation between the pressure in the reservoir, \( P \), and the flow rate through the die (exit flow rate), \( Q_{\text{ex}} \), an oscillatory flow is observed when the feeding flow rate or inlet flow rate, \( Q_{\text{in}} \), lies in the range \( Q_{\text{MFI}} - Q' \), as defined in figure 1. The amplitudes of the oscillations of the pressure and of the flow rate are respectively \( P_1 - P_2 \) and \( Q_1 - Q_2 \). The two functions pressure vs. time, \( P(t) \), and the flow

![Diagram of Exit flow rate vs. Pressure](image-url)
The second instability, or "sharkskin", occurs at flow rates, \( Q_{ss} \), lower than \( Q_{MFI} \). It consists of a regular surface pattern of ridges perpendicular to the direction of the flow. Naturally sharkskin means a baneful influence on the optical properties of the extrudates and is a limiting factor for several extrusion processes as film blowing or sheet die extrusion. Various mechanisms have been reported: cyclic build up and release of surface tensile forces, stick-slip, and differential recovery between the skin and the core (4–7, 15, 16).

The two phenomena are obviously periodic, but (i) the period of the former is about hundred times larger than that of the latter, 2 s and 0.02 s are typical; (ii) the amplitude of the oscillation of pressure of the former is about hundred times larger than that of the latter, \( 10^7 \) Pa and \( 10^5 \) Pa are typical.

It has currently been reported (1) that the physical characteristics of sharkskin differ so greatly from those of the MFI that these two phenomena probably have different physical causes. We assume this view to be incorrect. In this paper we first report on a phenomenological study of the pressure and output oscillations mentioned above (MFI) and then discuss these in terms of the relaxation oscillations theory. In the second part we show how a relaxation oscillator located at the die entry gives rise to a sharkskin phenomenon and we conclude that the two phenomena issue from the same physical origin, the double branched diagram, but in the one case the whole die vibrates whereas in the other only the die entry does.

2. Experimental

We used four experimental HDPE samples supplied by the Compagnie Franfaise de Raffinage, and one commercial product, HGF 4760, produced by Hoechst A.G. Their molecular characteristics are shown in table 1. Extrusion experiments were carried out at 190°C with a constant-speed capillary rheometer, ZWICK, type 7901.

<table>
<thead>
<tr>
<th>HDPE</th>
<th>1</th>
<th>2</th>
<th>3</th>
<th>4</th>
<th>5</th>
<th>HGF 4760</th>
</tr>
</thead>
<tbody>
<tr>
<td>M.I. (ASTM-190–2160)</td>
<td>1.77</td>
<td>1.04</td>
<td>1.12</td>
<td>0.54</td>
<td>1.80</td>
<td></td>
</tr>
<tr>
<td>( 10^{-3} ) Mn</td>
<td>27.9</td>
<td>24</td>
<td>29.5</td>
<td>16.6</td>
<td>11.5</td>
<td></td>
</tr>
<tr>
<td>( 10^{-4} ) Mw</td>
<td>14.9</td>
<td>13</td>
<td>15</td>
<td>21</td>
<td>13.5</td>
<td></td>
</tr>
<tr>
<td>Mw/Mn</td>
<td>5.3</td>
<td>5.4</td>
<td>5.1</td>
<td>12.7</td>
<td>11.7</td>
<td></td>
</tr>
<tr>
<td>( r_1/r_2 )</td>
<td>0.45</td>
<td>0.52</td>
<td>0.56</td>
<td>0.66</td>
<td>0.72</td>
<td></td>
</tr>
</tbody>
</table>

3. Main flow instability: low frequency

3.1. Critical conditions

Experiments are conducted in order to estimate the value of the pressure, \( P \), and the flow rate, \( Q \), for which the MFI occurs. We use different dies and barrels:
- die diameters: 0.5–1–1.5 mm,