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Polymer Melt Extrusion

Two Separate Ranges for Shear Flow Instabilities with Pressure Oscillations in Capillary Extrusion of HDPE and LLDPE

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SUMMARY

In polymer melt extrusion at constant piston rates, it is well known that linear polymers show a characteristic type of flow instability within a single range of flow rates. In this range, pulsations of the extrusion pressure occur in connection with typical distortions of the extrudate. When several HDPE and LLDPE samples were tested, it was found that one HDPE and one LLDPE sample showed two separate ranges of pulsating pressure in connection with characteristic distortions of the extrudate.

INTRODUCTION

In polymer melt extrusion through slits and capillaries different types of flow instabilities have been described in the literature [1,2]. There is evidence that such instabilities can arise (type A) in the die inlet region in front of the die entrance [1], (type B) within the die [1-10], or (type C) at the die exit [11]. Accordingly, extrudates show different, characteristic types of distortions depending both on the type of instability and the type of polymer.

The experimental investigation of the instability phenomena is usually performed with capillary rheometers at (i) constant extrusion pressure p or (ii) constant piston velocity. The total extrusion pressure p is mainly required for two energy portions: The pressure loss, p_0, in the die entrance region in front of the die and the pressure loss, $\Delta p = p - p_0$, within the die. For an experimental method of a further separation of p_0 into an elastic and a viscous portion, we refer to [12,13], and for a substantial treatment of the delicate exit pressure problem, we refer to Lodge and de Vargas [14].

With the constant piston velocity of mode (ii) the apparent shear rate $q_n = 4q/\pi R^3$ (q = volume flow rate, R = radius of the capillary) is kept constant. In the regions of instable flow, q_n is nothing but a reduced, nominal flow rate. For a long die (e.g. L/R = 60, L = length of the capillary) the measured extrusion pressure p is approximately equal to Δp. For a very short die, on the other hand, p approximates p_0. In the following, we restrict our discussion to the raw curves $p(q_n)$ measured with this long and one short capillary (L/R = 1). A more detailed analysis would require the Bagley-plot and the Rabinowitsch-Weissenberg correction which are of no relevance here.

Type B of the flow instabilities that arises within long dies, was first investigated for high density polyethylene (HDPE) [4,7], but it has also been observed in tetrafluoroethylene and hexafluoropropylene (TFE-HFP) copolymer [1] and linear low density polyethylene (LLDPE) [10]. For these polymers and mode (i) of constant pressure, the $q_n(p)$-curves for long capillaries show a step in q_n at a critical pressure which is higher for increasing p than for decreasing p, in other words the curve $q_n(p)$ has a hysteresis. During this step the shape of the extrudate changes from smooth to screwlike. For mode (ii) with constant piston velocity, for the same die and the same polymers, in a certain region of q_n pressure oscillations occur indicating instable flow. In contrast, for very short dies (e.g. L/R = 1), there is no step in $q_n(p)$ with mode (i) [15] and, consequently, the pressure should not oscillate in any region with mode (ii). Obviously, this type B of flow instabilities does not originate in front of the die but rather within the die. The same conclusion follows from birefringence studies [4].

Investigations of polymer melt flow instabilities were included in a recent research project of the IUPAC (Macromolecular Division) Working Party on Structure and Properties of Commercial Polymers. For two of the distributed polymer samples, the authors found two separate ranges of oscillating pressure with mode (ii) and a very long (but not with a very short) die. This result indicates two ranges of flow instabilities of type B.

EXPERIMENTAL

The measurements of p(q) were performed with mode (ii) of constant piston velocity in a capillary rheometer (RHEOGRAPH 2000, Goettfert, Germany) with two dies of 1 mm diameter and L/R = 1 and 60. The piston diameter was 15 mm. The pressure transducer (Dynisco) had a range of 10^8 N/m^2 and was calibrated by means of a hydraulic balance at the temperature of the measurements (190^{o}C).

For the detailed characterization of the two polymers reference is made to the forthcoming paper of the IUPAC Working Party. The trade names for the two samples are as follows:
HDPE: Alathon 7030 of DuPont
LLDPE: Lotrex FW 1290 of Chimie de France

RESULTS

Figure 1 shows the extrusion pressure p as a function of the apparent shear rate q_n. For the short capillary (lower curve) no instability could be observed. For the long capillary (upper curve) p increases continuously with q_n up to point A at which pressure oscillations start to occur until point B is reached. Between B and C no oscillations of the pressure occur. At point C a second range with pressure oscillations begins and reaches up to point D. At point D the final range begins with no pressure oscillations, p increases steadily as in the range B-C. The limits for all five ranges, the magnitude of the pressure oscillations, and the corresponding shape and surface of the extrudate are given in Table I. The extrudate surface is characteristic for each range of the upper curve of Fig. 1 (and Fig. 2). In passing we add that in spite of the constant piston velocity the output rate oscillates in the ranges with pressure

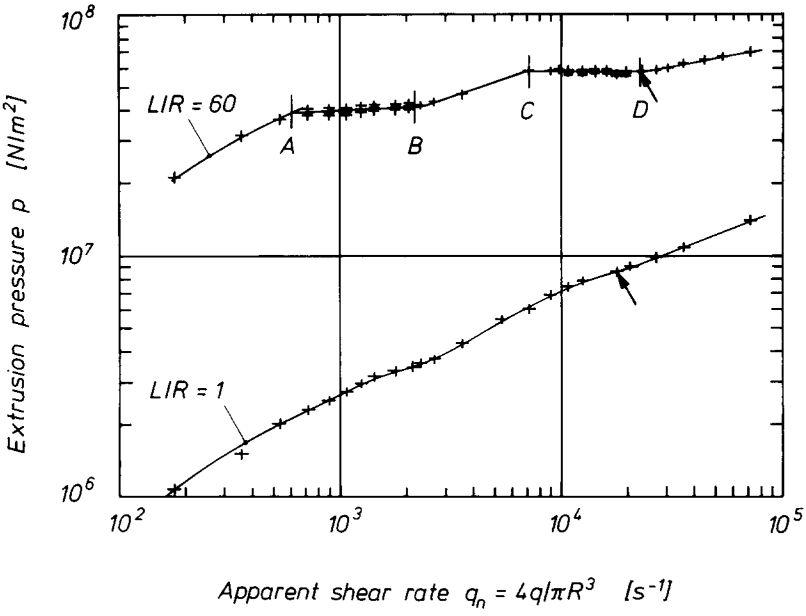

Figure 1: Extrusion pressure p as a function of the apparent shear rate $q_n = 4q/\pi R^3$ for the HDPE sample at 190°C. Capillaries: R = 0.5 mm, L/R = 60 (upper curve) and L/R = 1 (lower curve). Ɨ Maximum, average and minimum values of the pulsating pressure. The arrows indicate the onset of the type A instability arising in the die entrance region.

oscillations, and both oscillations, for q_n and p, have the same frequency.

The arrows in Figure 1 indicate the onset of a seriously distorted extrudate which occurs with both capillaries and, therefore, corresponds to the onset of flow instabilities of type A. Figure 2 shows the extrusion curve $p(q_n)$ for the LLDPE sample. The general result is the same as concluded from Figure 1 for the HDPE sample. Again the details are given in Table I.

CONCLUSIONS AND DISCUSSION

The main conclusion of this paper is that for one LLDPE and one HDPE sample out of three different HDPE's the extrusion behaviour with a long capillary shows two separate ranges of instabilities characterized by pressure oscillations when the extrusion is performed with constant piston velocity, and that these two regions are separated by a region without such oscillations. As Table I indicates, however, in this central region the surface of the extrudate is not completely smooth. On the other hand, in this region the slope of the $p(q_n)$-curve is similar to that of the first and the last range and remarkably different from the slope of nearly zero in the two adjacent ranges with pressure oscillations. Therefore, the flow mechanism in the central region must be different from that in the two instable regions with pressure oscillations. It is not clear whether there is a laminar flow in the central region as it is the case in the first region ($q_n < A$), or whether there

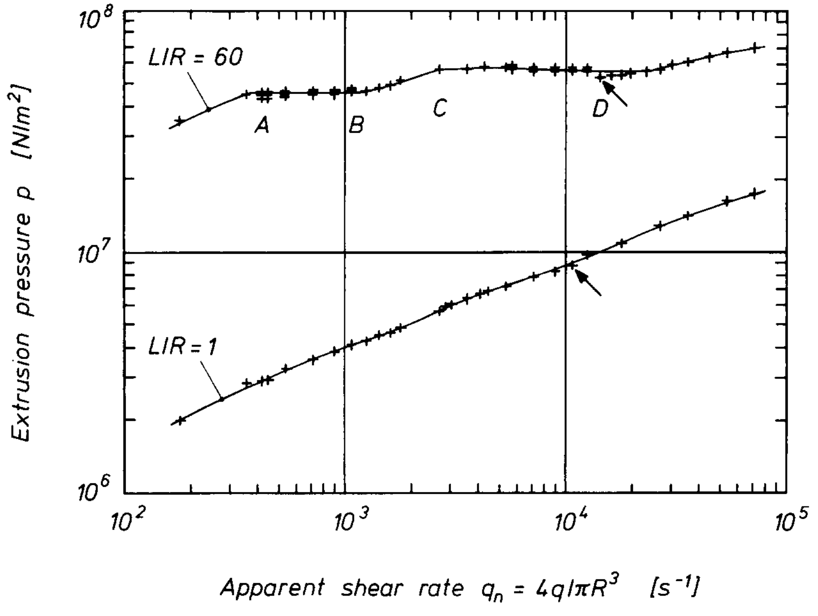

Figure 2: Extrusion pressure as a function of the apparent shear rate for the LLDPE sample. Test conditions and symbols as in Figure 1.

are already flow instabilities that are characteristic for the last range (q_n > D) in which no pressure oscillations occur but remarkable extrudate distortions. These distortions must have their origin in the die entrance region (type A of flow instabilities) because they are found also with the very short die.

Our finding of two ranges with pressure oscillations separated by one range without such oscillations is very strange: This effect does occur only in one HDPE sample but not in two other HDPE samples from different suppliers. It is not at all clear which structural features of the polymer are responsible for the effect described. Consequently, we cannot claim that this finding is true for LLDPE in general because we investigated only one sample of that type of polymer.

It should be mentioned that the occurrence of two instable flow regions with extrudate distortions and separated by a second region of stable flow with smooth extrudate was quoted in the literature [16-18], but was originally observed and only briefly mentioned by Ui and coworkers [15] for a polypropylene melt. It is an open question whether these instabilities, characterized by a distorted extrudate of a polypropylene sample, can be correlated with the instabilities characterized by pressure pulsations described here for the HDPE and LLDPE samples investigated.

Table I: Ranges of stable and instable flow of the HDPE and the LLDPE sample for the long die L/R = 60 and shape of extrudate. The points A...D refer to Figures 1 and 2.

	Range of apparent shear rate [s^{-1}]	Amplitude of pressure oscillations	Shape and surface of extrudates
HDPE	$q_n < A = 6.0x10^2$	no	smooth to matt
	$A < q_n < B = 2.1x10^3$	± 5 %	alternatingly smooth and screwlike
	$B < q_n < C = 7.1x10^3$	no	slightly distorted (near B) to screwlike (near C)
	$C < q_n < D = 2.2x10^4$	± 3 %	alternatingly smooth and screwlike
	$q_n > D$	no	seriously distorted
LLDPE	$q_n < A = 3.8x10^2$	no	smooth to matt
	$A < q_n < B = 1.2x10^3$	± 5 %	alternatingly smooth to screwlike
	$B < q_n < C = 2.7x10^3$	no	slightly distorted (near B) to screwlike (near C)
	$C < q_n < D = 1.4x10^4$	± 3 %	alternatingly smooth to screwlike
	$q_n > D$	no	seriously distorted

Concerning the physical reasons for the onset of melt flow instabilities it is not our intention to add one more speculation to the many already published in the literature. In our opinion only investigations with new methods can bring more insight. The laser-Doppler method of velocimetry, applied to polymer melt flow also in the instable regions, may probably be the experimental key for the final solution of this question.

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