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Flow of Polymer Melts Through a Well-Lubricated, Conical Die

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Synopsis

The concept of gross melt fracture of polymer melts as a tensile failure in the die entry region was supported in this work by the observation of a dramatic increase in the melt fracture of polyethylene extrudates upon lubricating thoroughly a conical, converging extrusion die. This flow, according to an analysis using a Fromm viscoelastic model, was found capable of producing axial tensile stresses in the extrudate in excess of 10^6 dynes/cm^2 at the very moderate exit shear rate (no lubricant) of 100 sec^{-1}. A calculated stress level of about 5×10^6 dynes/cm^2 caused sharp, deep transverse cuts to appear in the extrudate. The ability of tensile stresses of this magnitude to fracture melts was demonstrated by separate experiments run in simple tension on molten rods, using similar rates and total deformations. A large qualitative difference between high- and low-density polyethylene in both these experiments was noted.

INTRODUCTION

Recently, there has been considerable interest in extensional flows of polymer melts, both as a tool for checking constitutive descriptions[1,2] and because of the importance of this deformation mode in the fabrication of plastic articles.[3,4] It has been found that a flow emphasizing extension, but with elements of shear as well, can be easily and reproducibly generated by lubricating a conical, converging die. The flow thus probably resembles the convergence of melt into a die, but with a controlled angle. The most evident qualitative feature of this flow is the early onset of gross melt fracture, supporting the observations of Ballenger and White[5] for the flow of melts into a die at high rates and the ideas of Everage and Ballman[6] concerning the origin of gross melt fracture in the die entry region.

ANALYSIS

If it be assumed that the converging, conical die can be perfectly lubricated, reducing the wall shear stresses to zero, the equation of motion in spherical coordinates simplifies immensely, leaving

$$\frac{\partial p}{\partial r} = -\frac{3\tau_{rr}}{r} - \frac{\partial \tau_{rr}}{\partial r} \qquad (1)$$

where p is pressure, r is the radial coordinate, and τ is stress.

The further assumptions have been made that inertial and body forces are negligible and that the material obeys a constitutive description with

$$\tau_{rr} = -2\tau_{\theta\theta} \tag{2}$$

$$\tau_{\theta\theta} = \tau_{\phi\phi}. \tag{3}$$

Fluid mass continuity provides the following equation:

$$\dot{\gamma} = \begin{pmatrix} 2 & & \\ & -1 & \\ & & -1 \end{pmatrix} \frac{2c}{r^3} \tag{4}$$

where $\dot{\gamma}$ is the rate of deformation tensor and c is a constant equal to the flow rate Q divided by $2\pi(1 - \cos\theta_0)$; θ_0 is the half-angle of entry (Fig. 1). It can be seen from eq. (4) that the flow is a simple extensional process with rapidly increasing rate. Clearly, the complete absence of vorticity even under conditions of perfect lubrication is unlikely, particularly around the entrance and exit and even in the die itself should surfaces of equal velocity choose to be anything but spherical shells. But it is felt that the primary contribution to pressure loss is due to extensional deformation.

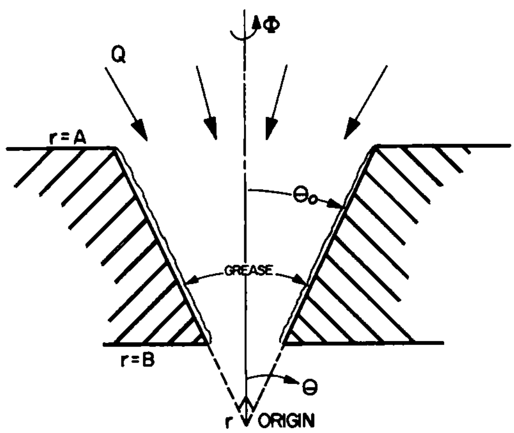

Fig. 1. Die geometry in cross section.

To derive the pressure from the equations of motion, it is necessary to describe the fluid properties. Because an estimate of tensile stress and its relationship to fracture phenomena was of primary interest, a simple viscoelastic model was used to derive a relationship between total pressure drop and maximum tensile stress. The tensile stress should be a maximum at the die exit, with the total stress being roughly comparable with the dynamic stress, because the pressure is near zero. By expressing stress in terms of pressure drop instead of material parameters, one has a reasonable assurance of a fair degree of freedom from the shortcomings of the model:

$$\tau_{ij} + \lambda \frac{\mathfrak{D}\tau_{ii}}{\mathfrak{D}t} = -\eta\,\dot{\gamma}_{ij} \tag{5}$$

where λ is the fluid's characteristic time, η is the viscosity, and $\mathfrak{D}/\mathfrak{D}t$ is the

Jaumann derivative. This model is consistent with the assumptions in eqs. (2) and (3).

The solution of this model for the assumed conditions is

$$\tau_{rr}(R) = e^{R^3/3C\lambda} \frac{4\eta}{\lambda} \int_A^R \frac{1}{r} e^{-r^3/3C\lambda} \, dr \tag{6}$$

with a (positive) pressure drop given by

$$\Delta P = \tau_B + 3 \int_A^B \frac{\tau_{rr}(r)dr}{r} \tag{7}$$

where A and B are the initial and final radii, respectively. Solution of ΔP in terms of τ_B, the maximum stress, was done numerically, with checks by asymptotic solutions at low and high rates. Dependence of the function $\Delta P/\tau_B$

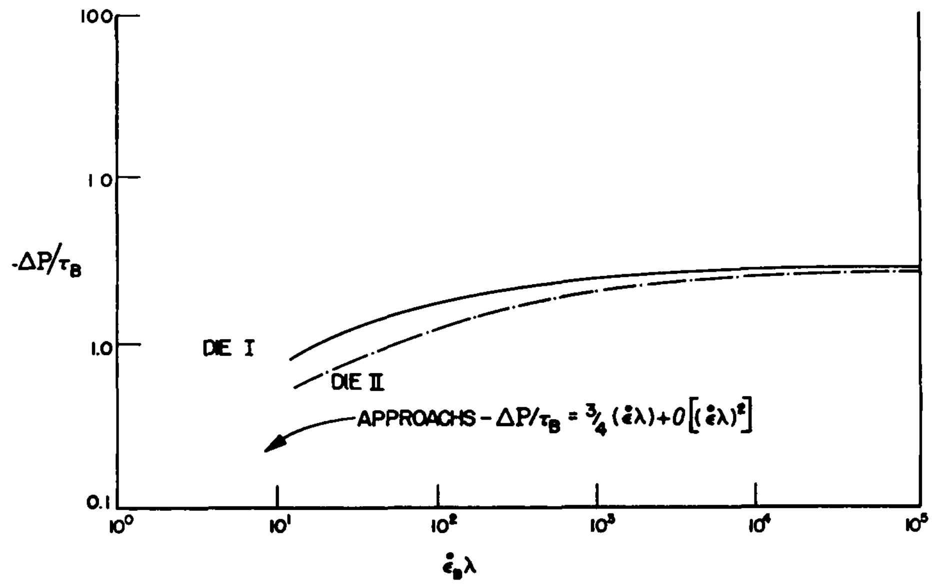

Fig. 2. Fromm model predictions for lubricated conical die.

on material constants and flow conditions is small, as can be seen in Figure 2. The high-rate "elastic" solution is

$$\tau_B = \frac{\Delta P}{\frac{3}{2} \ln \frac{B}{A} + 1} \tag{8}$$

corresponding to a stress–strain relationship

$$\tau_{rr}(R) = \frac{4\eta}{\lambda} \ln \frac{R}{A} \tag{9}$$

and gives the lowest exit stress per unit pressure drop available with the Fromm model under the assumed kinematics.

EXPERIMENTAL

Materials

The sample designated LDPE was a 0.2 melt-index, 0.915-density polyethylene, while the high-density polyethylene sample (HDPE) had a melt index of 0.3 and a density of 0.955.

Rheometer and Procedure

A constant flow-rate piston rheometer with a 2.5-cm bore was loaded with pellets and equilibrated for 15 min at 190°C, maintaining a low extrusion rate through a capillary die. Without disturbing the melt, the capillary die was removed and replaced by a conical die (Table I), well coated with grease. After a few minutes of equilibration, the extrusion was continued at a fixed flow rate, and pressure was monitored. Extrudates were saved.

TABLE I
Die Geometries

Die no.	A, cm	B, cm	θ_0, degrees	Total deformation ratio
I	1.45	0.11	60	173
II	1.98	0.17	40	136

RESULTS AND DISCUSSION

As expected, the grease lowered the pressure relative to a clean die, and the pressure gradually increased as the grease wore away. Of the greases tried,

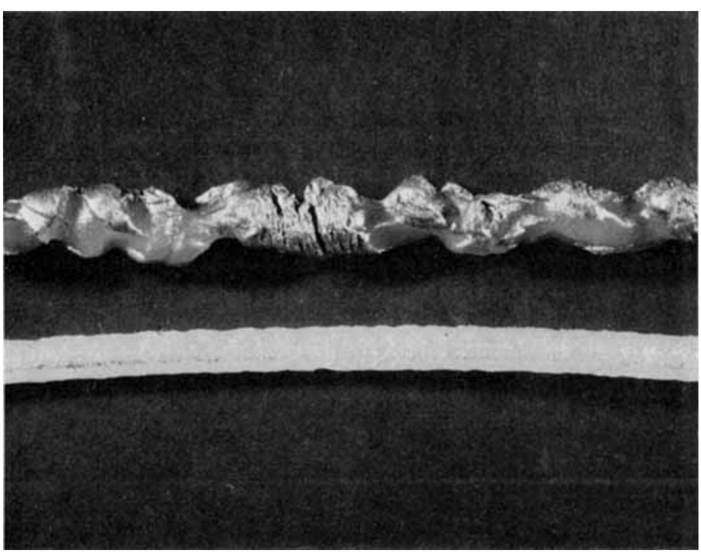

Fig. 3. Extruded strands, LDPE, 190°C, 0.07 cm^3/sec, die I. Upper strand: extrudate from die greased with "Never Seeze" lubricant, which is visible on the outer surfaces and absent in the deep, transverse cuts. Lower strand: extrudate after most lubricant has worn off die walls. Deep cuts are absent, die swell is reduced.

TABLE II
Pressure Drops, Maximum Stress, and Extrudate Condition Using Conical Dies Lubricated with Silicone Grease

Die no.	Resin	Q, cm^3/sec	ΔP, (dynes/cm^2) $\times 10^{-6}$	τ_B, (dynes/cm^2) $\times 10^{-6}$	Extrudate appearance
I	LDPE	0.007	6.2	4.8	smooth
		0.028	10	5.1	some roughness
		0.07	18	8.2	very rough, cuts
	HDPE	0.028	7.0	3.7	quite smooth[a]
		0.07	13	5.9	a few cuts
		0.49	26	10	many deep cuts
II	LDPE	0.007	3.9	5.8	smooth
		0.028	6.6	5.9	very rough, cuts
	HDPE	0.07	8.3	5.5	quite smooth[a]
		0.28	14	7.4	quite smooth[a], with occasional cuts

[a] Some "matte" appearance on surface.

all produced about the same pressure decrease, but silicone grease was more durable than "Never Seeze" or "Molycote."

The important result was the melt fracture. In spite of *lower* pressure drops in the greased die, the fracture ("gross distortion" variety) was *worse* (Fig. 3). The grease, according to the assumptions made here, forces the melt to converge at the sharp angle of the die itself, rather than establishing a more gradual convergence dictated by a balance of shear and extensional forces in the entry region.[7] By eliminating shear stress, the grease has tipped the balance in favor of extension, generating high tensile stresses. The resulting melt fracture, as will be shown, can be logically assigned to these stresses.

Miller[8] has shown that $\lambda = 10$ sec is a realistic value for polyethylenes of this grade, implying that the magnitude of $\dot{\epsilon}_B\lambda$ will be greater than 100 and well into the region of fairly constant $\Delta P/\tau_B$ in Figure 2, for most of the flow rates used. Using $\lambda = 10$ sec, the values for the maximum tensile stress τ_b

TABLE III
Tensile Failure of Polyethylene Melts

		Values at break		
Resin	Experiment	Time, sec	Stress, (dynes/cm^2) $\times 10^{-6}$	Extension ratio
LDPE	lubr. die I	11	5.1	173
	II	14	5.9	136
	tensile	120	2.6	43
		18	6.5	33
		2.5	12	23
HDPE	lubr. die I	5	5.9	173
	II	1.4	7.4	136
	tensile	53	(0.6)[a]	17
		0.8	(9.8)	10
		0.12	(120)	24

[a] Ill-defined failure; see text.

were taken from Figure 2 for each flow rate and listed in Table II. That these stresses can reasonably produce fracture was demonstrated by tensile experiments in an apparatus similar to that described by Chen et al.[4] Failure parameters are listed in Table III for this increasing stress experiment, along with estimates from Table II for the lubricated die results. It is clear that the tensile stress developed in the conical die is high enough to produce fracture. The times are given to demonstrate that the deformation processes were of roughly comparable rates.

As described by Chen et al.,[4] it was difficult to draw uniformly the HDPE in a weight-drop experiment. By concentrating upon temperature uniformity and stability, data such as those in Table III could be obtained. It should be noted that the HDPE samples, in spite of these precautions, always broke by a flow process even at very high stresses. On the other hand, the LDPE samples, subjected to stresses of greater than 10^6 dynes/cm^2, invariably failed by a crack propagation mechanism. In some instances, *two* cracks were observed in a single sample. This qualitative difference is also disclosed by the relative ease of producing gross melt fracture in the greased conical die and the more rounded cuts in the HDPE strands when fracture did occur.

The differences between low- and high-density polyethylene melts in failure—stated without demonstration at this time—are explainable in terms of the differences in their isochronous stress–strain behavior: linear polyethylene appears to have a stress maximum whereas branched polyethylene produces elastomer-like stress–strain relationships. A reasonable qualitative guess at molecular reason for this would be a knotting, upon straining, of chain entanglements involving molecules with medium or long branches.

This simple experiment points out that die design for minimum pressure drop is almost by definition incompatible with smooth extrudates because such a design must invariably increase axial extension rates. Furthermore, minimizing shear deformation and its derivatives in die design may cause *higher* tensile stresses for many shear sensitive fluids even if the tensile deformation history could be held constant. As the experiments explicitly demonstrate, the use of effective external lubricants with a given converging die cannot be expected to alleviate distortion problems for both of the reasons stated above.

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