LETTERS TO THE EDITORS

Filament Distortion and Die Entry Angle Effects in Polyethylene Extrusion

Visual evidence has been presented by various authors[1–6] showing the phenomenon of extrudate distortion in the capillary extrusion of elastic melts like polyethylene to be associated with a melt fracture occurring near the entrance to the capillary die. As yet no satisfactory mechanism has been suggested to explain why such visual extrudate distortion should accompany melt fracture. Further, several authors[1,3,7,8] have reported evidence which seems to show a dependence of the critical stress for melt fracture, τ_d, on the geometry at the die entry of conically tapered dies, the stress becoming greater as the included die entry angle diminishes.

The purpose of this communication is to discuss the cause of extrudate distortion during melt fracture, and to elaborate on the dependence of τ_d on die entry geometry.

Following Spencer and Dillon[6] and Schulken and Boy,[9] we regard the flowing polymer melt as a sink for the absorption of applied deformation energy. As part of a general theory of polymer flow, to be published separately, this capacity for storing deformation energy is concluded to be limited and defined by the size and shape of the constituent chains of a given polymer melt. Melt fracture is therefore regarded as an additional means of dissipating applied energy which is actuated when the capacity of the melt for recoverably storing applied deformation energy becomes exhausted. This concept is basic to the ensuing considerations.

1. The Nature of Extrudate Distortion

Bagley and Birks[5] have shown that the extruding polyethylene forms a characteristic flow pattern within the extrusion device. The shape of the flow pattern depends on structural properties of the melt. Figure 1, which is based on Figures 2 and 3 of reference 5, shows typical flow patterns for a linear (A) and a branched (B) polyethylene. The flowing branched polyethylene creates a distinct dead space within which only melt circulation occurs. On the other hand, the flow lines in the linear melt are associated with virtually no dead space.

Consider now a region that is near the entrance to the capillary orifice in the flowing polymer B. As the applied stress increases, the population within this region becomes progressively more elastically extended. Simultaneously, increasing normal forces continually compress the circulating melt within the dead space. When the potential for storing deformation energy is exhausted, excess applied energy is dissipated in the flowing melt through a fracture mechanism, and entry of some dead space material into the flowing polymer stream occurs. The injected material is, however, much less elastically extended and much more viscous than the flowing melt. Since melt from the dead space enters near the capillary orifice,[5] its dwell time in the flowing stream is short, and it cannot be elastically deformed to the same state as the flowing melt under the given conditions of applied stress. The extrudate can consequently be viewed as a mixture of melt components with radically differing potentials for elastic recovery, thereby accounting for the pronounced distortion effect[8] in the extrudate during extrusion beyond the melt fracture stress. This mechanism suggests that the views of Tordella[1,2] and Spencer and Dillon[6] on the nature of the distortion phenomenon are in a sense compatible. Further, on the basis of this argument, the die length should affect the severity of extrudate distortion.[10] At any constant shear rate, the dwell time of a unit volume of melt in the capillary increases with the length of that capillary. More time is therefore available for the melt constituents to become elastically homogeneous. The severity of visual distortion may then be expected to diminish with increasing die length. The increase in the degree of extrudate distortion as the stress rises above τ_d can be attributed in part to the rising output rate, as a result of which the dwell time of a unit melt volume in the capillary decreases, and, initially, to the increasing frequency of melt fractures. Conceivably, however, a continuous increase in the frequency of melt fractures may eventually decrease the amount of extrudate distortion. At some stress well above τ_d the fracture frequency may become so high as not to allow sufficient time for significant surges of melt from the dead spaces. Moreover, at these high stresses, the dead space tends to disappear.[5] Melt fracture then becomes accompanied by surges from regions in which the melt is more elastically akin to the melt in the center of the flow funnel. Consequently, the extrudate now becomes elastically more homogeneous. These considerations suggest that the visual distortion of the extrudate passes through a maximum in the stress region above τ_d.

The extrusion of an elastically heterogeneous extrudate then does not depend on the presence of a dead space alone. Indeed, extrudate distortion is noted when, at stresses well above τ_d, the dead space disappears. As suggested above, elastic heterogeneity in the extrudate can be obtained whenever specific regions containing melt with different amounts of stored elastic energy take part in flow. Now, the stored elastic energy of a flow element decreases from a maximum at the center of the flow stream to the wall of the extrusion device (see Fig. 1). Surges of melt from the less elastically deformed regions which accompany melt fracture, also constitute a mechanism for creating extrudate heterogeneity. Moreover, flow elements involved in a retraction following melt fracture[5] may be considered as elastically relaxed in comparison with their surroundings. Thus, at some fracture frequency, sufficient elastic heterogeneity for gross visual distortion could be achieved without surges of dead space melt. This is, of course, the situation in the extrusion of linear polymer melts. At τ_d, when melt fracture begins, extrudate heterogeneity is due almost entirely to surges of melt from the flow regions of lower stored elastic energy. One would not expect this heterogeneity to be as severe as that caused by injection of dead space melt, and therefore the initial visual effects of melt fracture should be slight.

Violent extrudate distortion can be noted in linear polyethylene melts, however.[11] This would seem to be associated with a sufficiently high fracture frequency to compensate for the gross effects of a dead space region. Such a frequency of melt fractures would tend to occur at stresses well above the true τ_d. The argument suggests that values of τ_d determined from the onset of gross extrudate distortion are higher than the true stress at which melt fractures commence.*

2. Dependence of τ_d on Die Entry Geometry

Since melt fracture is associated with the exhaustion of a potential for storing deformation energy within the flowing melt, then the melt fracture stress τ_d is a function of the size and shape of the chains constituting a given melt, but is entirely unrelated to the geometry at the entry of a given extrusion orifice. The reported effects of die entry taper[1,3,7,8] are therefore attributable to the influence die taper has on the aftereffect of melt fracture, that is, the degree of elastic heterogeneity in the extrudate.

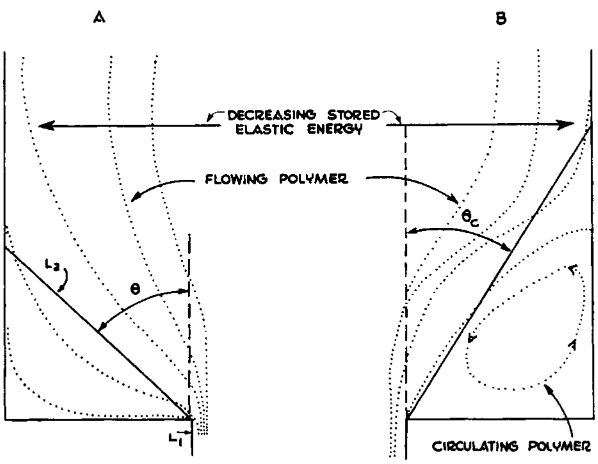

Fig. 1. Flow patterns formed during capillary extrusion of (*A*) linear and (*B*) branched polyethylene (after Bagley and Birks[5]).

Compare the extrusion of a branched polyethylene B (Fig. 1) through a flat entry die and a die of the same length to radius ratio as the flat entry, but with an included entry angle θ_c which just eliminates the dead space created by the natural flow lines of the polymer. The onset of melt fracture is now marked by surges of flowing melt from regions in which the stored elastic energy is more nearly equal to that associated with the main flow stream. Thus, although the critical stress for melt fracture should be invariant in the two cases, the reduced visual aftereffects of melt fracture in the tapered die extrusion will indicate an apparent rise in τ_d. Analogous with the above arguments, gross visual effects of melt fracture should now become apparent only at a stress considerably above the true τ_d value. The independence of τ_d and die entry taper has been established experimentally, and is discussed in more detail in a forthcoming publication.[12]

A further die entry effect must be recognized if the die taper interferes with the naturally formed flow lines of the polymer. With a linear polyethylene (A) (Fig. 1), this is the case whenever the total included die entry angle 2θ is less than 180°. Tapering now may lead to excess pressure losses in the conical section above the die entry. A convenient expression for these pressure losses in terms of an increase in the effective die length L_e may be written as

$$L_e = L_1 + L_2 f(\theta) \quad (1)$$

where L_1 is the effective land length of the die, L_2 is the length of the tapered wall, and $f(\theta)$ is some function of the included entry angle. Equation (1) is also applicable to branched polyethylene extrusion when $\theta < \theta_c$ the difference $(\theta_c - \theta)$ would, however, be employed. Thus, even though τ_d is again invariant, there may be a real increase in the pressure at which melt fracture begins as the included angle diminishes.

* One interesting method of determining the true τ_d involves a measure of the thermal effects which should accompany the release of stored elastic energy on melt fracture. Temperature conditions in the extruding melt are being studied in this laboratory, and results will be discussed in a future publication.

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