Tear strength of polyethylene

D. -S. CHIU, A. N. GENT*, J. R. WHITE
Institute of Polymer Science, The University of Akron, Akron, Ohio 44325, USA

The fracture energy of moulded sheets of polyethylene has been found to depend strongly upon the thickness of the sheet, increasing linearly over the range 0.05 to 1.0 mm. This variation is attributed to a dependence of the volume of the plastic zone at the crack tip upon t^2, where t is the torn thickness. By extrapolation, threshold values of fracture energy were determined at zero thickness. These represent the strength in the absence of large-scale plastic yielding. The fracture energies of both thin and thick sheets of high-density polyethylene (HDPE) were found to vary strongly with rate of tearing and test temperature, passing through maxima at particular rates and temperatures. This behaviour is attributed to corresponding changes in local ductility. A general correlation was found between the dependence of fracture energy upon tear rate and the dependence of loss modulus E'' upon the frequency ω of small oscillatory deformations. It is concluded that the work of fracture is mainly expended in local yielding at the tear tip, on the scale of single spherulites, about $4\,\mu m$ for HDPE. Annealing HDPE at $120°$ C reduced the fracture energy of thick sheets to about $10\,kJ\,m^{-2}$ but did not greatly alter the threshold strength. In contrast, annealing sheets of LDPE had relatively little effect on the fracture energy at any thickness.

1. Introduction

Surprisingly little information has been published on the tear strength of semicrystalline polymers although considerable attention has been paid to yielding and rupture in tension; see, for example, [1–10]. As far as the present authors are aware, the first measurement of the energy required to fracture polyethylene by tearing was carried out by Anderton and Treloar [11]; they examined the effect of orientation produced by drawing upon the tear strength of low- and high-density polyethylene, and in the course of this work obtained values for the fracture energy of undrawn materials, of about $160\,kJ\,m^{-2}$ and $77\,kJ\,m^{-2}$, respectively. In a closely similar study of polypropylene, Sims found a value of about $120\,kJ$ m^{-2} at $20°$ C, with larger values at higher temperatures and lower ones at lower temperatures [12]. These fracture energies are considerably larger than those for glassy polymers, typically about 0.1 to $1\,kJ\,m^{-2}$ [13] or even those for filler reinforced elastomers, about 10 to $30\,kJ$ m^{-2} [13], reflecting the well-known toughness of films of semicrystalline polymers.

A number of questions remain unanswered, however. Are these tear energies strongly dependent upon the rate of tearing, as well as upon the temperature of test, as would be expected for viscoelastic materials? Are the measured values characteristic of the material or do they depend upon the particular test method used to determine them? Anderton and Treloar found that different test methods gave similar values of tear energy for highly-oriented, relatively brittle materials, but they reported "inconsistent" results for unoriented, ductile materials [11]. Moreover, Anderton and Treloar [11] and Sims [12] used unoriented films of a single thickness (0.6 and 0.3 mm, respectively) in their experiments, although Vincent has shown that the fracture energy of unoriented polyethylene terephthalate, which somewhat resembles polyethylene in ductility,

depends directly upon the thickness of the film [4]. Indeed, Isherwood and Williams [14] found a direct proportionality between the fracture energy (work expended per unit area torn through) and the thickness of the test sheet for a wide range of ductile materials, including a polyethylene sample. An exploratory study has therefore been carried out of the effects of film thickness and of the rate of tearing and test temperature, upon the fracture energy of moulded sheets of several types of polyethylene. The results are presented here.

2. Experimental procedure
2.1. Materials
Three polyethylene resins were used in this research: a low-density (LD) film grade polyethylene, having approximate molecular weights $\bar{M}_n = 1.9 \times 10^4$ g mol^{-1} and $\bar{M}_w = 1.2 \times 10^5$ g mol^{-1} and a density at room temperature of 0.925 Mg m^{-3}; a medium-density (MD) detergent-grade polyethylene copolymerized with a few per cent of 1-hexene, having approximate molecular weights $\bar{M}_n$ of 2.5×10^4 g mol^{-1} and $\bar{M}_w = 1.6 \times 10^5$ g mol^{-1} and a density at room temperature of 0.95 Mg m^{-3}; and a high-density (HD) linear polyethylene having an approximate molecular weight $\bar{M}_w$ of 1.2×10^5 g mol^{-1} and a density at room temperature of 0.965 Mg m^{-3}. The LD polymer was supplied by Exxon Chemical Company and the other two polymers were supplied by Phillips Chemical Company, denoted Marlex HHM 5202 and Marlex EMB 6035, respectively.

Thin sheets of polyethylene were prepared from the original granules by moulding for one hour at a temperature of 150° C. After this period the metal mould was placed in a warm water bath at about 45° C to cool the sheet quickly. This procedure is termed "quenching". Some sheets were then reheated to 110 or 120° C for a period of 1.5 h and then cooled slowly back to room temperature at a rate of about 1° C min^{-1}. These sheets are termed "annealed".

Some physical properties of the test materials are listed in Table I. The fractional degree of crystallinity C was calculated from the measured density ρ, using a relation based on that given by Chiang and Flory [15, 16]:

$$0.145 \, C = 1 - (0.855/\rho)$$

The yield stress σ_y, referred to the original cross-sectional area of the testpiece, the breaking elongation e_b, and the work U_b required to break the test-piece per unit volume, calculated from the area under the force-displacement curve up to the breaking point, were all measured using small dog-bone-shaped test-pieces having an effective length and width of about 4 mm.

Annealing was found to increase the density and degree of crystallinity only to a small extent in all cases, and the tensile properties of LDPE did not change significantly. For HDPE, however, (and to a lesser extent for MDPE) the breaking elongation and work-to-break were drastically reduced by annealing. Indeed, HDPE changed from a tough, ductile material to a relatively brittle material. This change probably reflects a significant decrease in the number of interlamellar and interspherulitic tie molecules as a result of annealing HDPE [17].

2.2. Test methods
Tear test-pieces, 120 mm long and 20 mm wide, were cut from the moulded sheets and the time-average value of the tear force F determined for tear propagation at a constant speed (Fig. 1a). The rate R of tear propagation was varied over a wide range, 1×10^{-8} m sec^{-1} to 1×10^{-2} m sec^{-1}, and the test temperature was varied

TABLE I Properties at 25° C: density ρ, crystallinity C, tensile yield stress σ_y, breaking elongation e_b and work-to-break U_b per unit volume

Material	ρ (Mg m^{-3})	C (%)	σ_y^* (MPa)	e_b^*	U_b^* (MJ m^{-3})
LD (Quenched)	0.9225	50.5	10.5 ± 1	7.5 ± 2	72 ± 15
LD (Annealed)	0.927	53.5	12.0 ± 0.5	7.5 ± 2	70 ± 15
MD (Quenched)	0.9375	61	22 ± 1	7.3 ± 0.5	145 ± 20
MD (Annealed)	0.947	67	24.5 ± 1	4.8 ± 0.5	95 ± 20
HD (Quenched)	0.953	70	24 ± 3	7.8 ± 1	150 ± 20
HD (Annealed)	0.966	79	29 ± 1	1.0 ± 0.3	30 ± 10

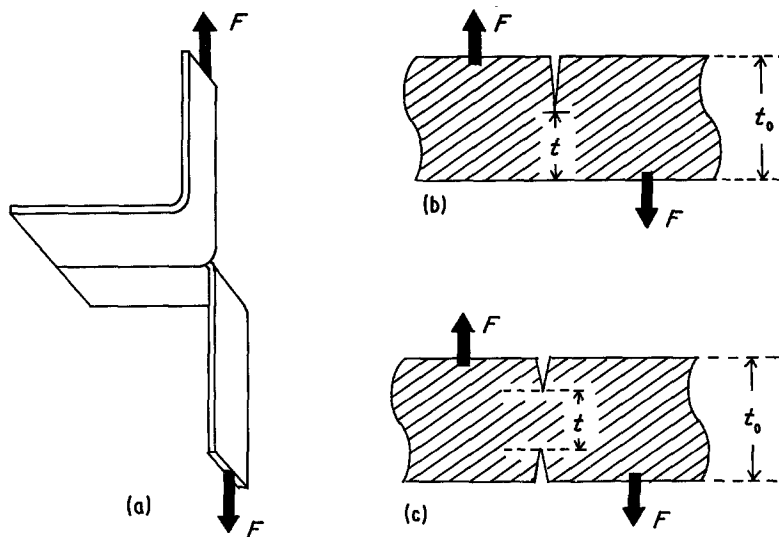*Measured at a strain rate $\dot{e}$ of 0.02 sec^{-1}.

Figure 1 (a) Tear test-piece I. (b) Cross-section of a single-grooved test-piece. (c) Cross-section of a double-grooved test-piece.

between room temperature, about 25° C, and 100° C.

In order to restrain the tear from deviating from a linear path and to minimize plastic yielding at the tear tip, deep grooves were cut in one or both surfaces of the test-piece parallel to the direction of tearing, as shown in the test-piece cross-section in Figs. 1b and c. Thus, the thickness t actually torn through was only about one-half of the original sheet thickness t_0. Moreover, as shown schematically in Fig. 2, the torn surface for single-grooved test-pieces lay generally at an angle of about 30° to the thickness of the sheet, probably because failure tended to occur at right angles to the principal tension stress. Values of the tear path width t were calculated
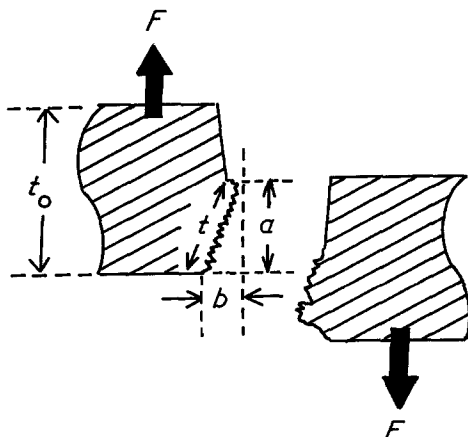

Figure 2 Sketch of torn surface of a single-grooved test-piece.

in these cases from measurements of the projected distances a and b, Fig. 2. No allowance was made for small-scale irregularities of the torn surface, which generally showed extensive plastic deformation anway.

Values of the fracture energy G_c were calculated from the tear force F using the relation [11, 18]

$$G_c = 2F/t \qquad (1)$$

i.e. assuming that energy expended in bending or stretching the test-piece legs was small in comparison with that expended in the vicinity of the tear tip. Attempts to measure energy expended in the test-piece legs indicated that it was, indeed, a relatively small fraction of the total fracture energy.

A second method of measuring tear energy is shown schematically in Fig. 3. Specially moulded cleavage test-pieces were used having the cross-section shown in Fig. 3b. In this case the width t of the tear path was determined by the distance separating two thin metal foils forming part of the mould, and the thickness h of material subjected to high stresses during tearing was determined by the thickness of the metal foils. Both the width t and the thickness h could be varied by suitable variations in separation and thickness of the metal foils. The thickness h was generally held small, however, about 75 μm, compared to the width t which was varied over the same range as the tear path width for the former test-piece (Fig. 1), from about 0.05 mm to 1 mm.
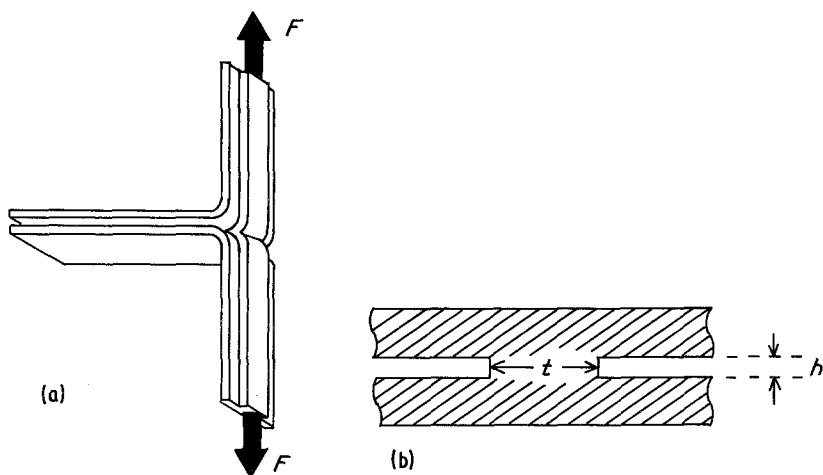
Figure 3 (a) Tear test-piece II. (b) Sketch of test-piece cross-section.

The fracture energy was again calculated by means of Equation 1 where t again denotes the tear path width, i.e. the width of the web of polymer connecting the upper and lower polymer films (Fig. 3b).

3. Experimental results and discussion
3.1. Effect of film thickness for type I test-pieces

The tear force F was found to increase approximately in proportion to the square of the width t of the tear path, instead of in direct proportion to t as would be expected for a simple fracture process. Thus, the fracture energy G_c from Equation 1 depends strongly upon t, almost in direct proportion.

A typical experimental relation between G_c and t is shown in Fig. 4. From such linear relationships, extrapolation can be made to zero width, yielding a threshold value G_0 of the tear energy

for a test-piece of infinitesimally small width. Also, values of G_c at an arbitrarily chosen width of 0.6 mm may be taken as representative of sheets of moderate thickness. They are denoted G_m. Thus, from measurements of tear energy for test-pieces having a wide range of thickness, and hence tear path width, values of G_0 and G_m appropriate to very thin and moderately thick test-pieces, respectively, were determined over a wide range of test conditions. They are discussed later. In the meantime, further attention is given here to the effect of film thickness upon the observed fracture energy.

A similar strong dependence of tear energy upon film thickness was found by Vincent [4] in experiments on polyethylene terephthalate films. He obtained a linear relationship with an intercept G_0 of $50 \, kJ \, m^{-2}$ and a slope dG/dt of $250 \, MJ \, m^{-3}$ over the thickness range 0 to 0.8 mm. For comparison, the results obtained here for polyethylene
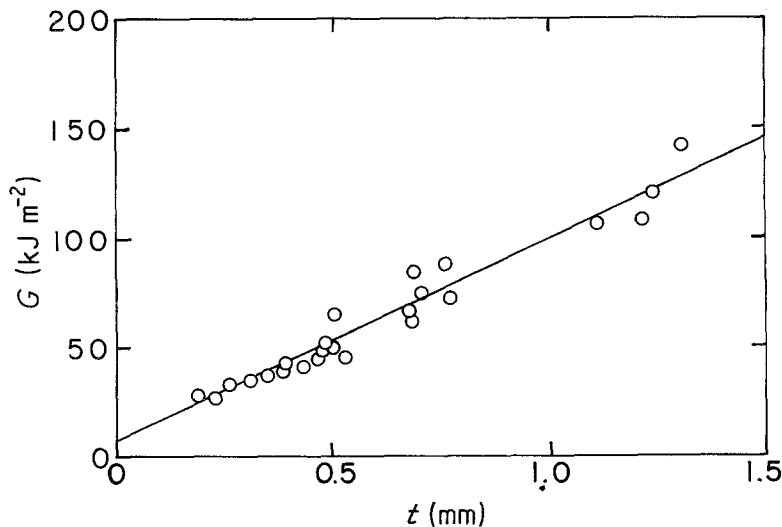

Figure 4 Experimental relations between tear energy G and tear path width t for Type I single-grooved test-pieces of quenched HDPE. Tear rate $= 0.4 \, mm \, sec^{-1}$, temperature $= 75°C$.

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vary from 2 to $10 \mathrm{kJ}\,\mathrm{m}^{-2}$ for G_0 and from 40 to $90\,\mathrm{MJ}\,\mathrm{m}^{-3}$ for dG/dt over the same range of thickness. Kambour and Miller reported a similar linear dependence of tear energy upon the width of the tear path for another ductile polymer; bis-phenol A polycarbonate [19, 20]. Thus, as Vincent surmised, this pattern of dependence upon thickness appears to arise from plastic yielding and flow at the tear tip, which takes place to an increasing extent with increasing thickness. It is in sharp contrast to the dependence of tensile strength upon thickness of test specimen, where a strong decrease is commonly observed for ductile materials. This is attributed to a change from fracture under plane stress conditions for thin samples to plane strain conditions for thick samples. The latter process takes place in a brittle way, at lower stresses and lower fracture energy [9, 21–23].

The observed tear energy G_c may be regarded as composed of two parts: G_0 arising from dissipation processes in the immediate vicinity of the tear tip, and an additional term G_p arising from plastic deformation in a larger region around the propagating tear. For simplicity, this second contribution to the fracture energy is assumed to be expended in tensile rupture of material having a cross-sectional area given by the product of the tear path width t and a dimension d in the perpendicular direction. The energy G_p expended per unit width of tear path and per unit distance torn is then given by

$$G_p = dU_b \qquad (2)$$

where U_b denotes the work-to-break in simple extension. A similar relation was proposed by Thomas for the fracture energy of rubber sheets,

with d denoting the effective diameter of the tear tip [24].

From the linear relations found to hold for polyethylene between G_p and the tear path width t, it appears that the dimension d is itself proportional to t in this case. Indeed, by comparing the experimentally determined slopes dG/dt with values of U_b, the ratio α of the effective diameter d of the plastic zone to the tear tip width t can be calculated from Equation 2.

Measurements were made of work-to-break U_b using small tensile test-pieces stretched at the same temperatures and at approximately the same rates $\dot{e}$ of elongation as the materials undergoing tear. The relationship assumed to hold between the rate R of tear propagation and rate $\dot{e}$ of elongation at the tear tip was: $\dot{e} = R/t$. Values for U_b obtained in this way are given in Table II.

Over a wide range of test conditions, leading to quite varied values of tear energy G, values calculated for d from Equation 2 were found to be generally similar to the torn thickness or tear path width t, Table II. Thus, the amount of material undergoing plastic flow and rupture as the tear advances by unit distance has a width t from geometrical considerations and a dimension in the perpendicular direction also approximately equal to t.

The magnitudes of d and t would be expected to be similar for yielding materials, from St. Venant's principle, as shown schematically in Fig. 5. In agreement with this hypothesis, values of dG/dt, and hence d, for double-grooved test-pieces were found to be smaller than those for single-grooved test-pieces by about 25 per cent (Table II). In these cases plastic flow is prevented

TABLE II Effective diameter d of plastic zone in HDPE, relative to width t of the tear path, calculated from Equation 2. Tear rate $= 40\,\mu\mathrm{m}\,\mathrm{sec}^{-1}$

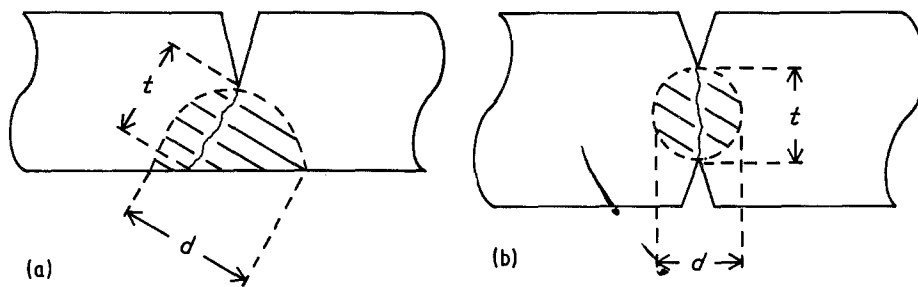| Temperature (°C)	U_b (MJ m⁻³)	Single-grooved test-pieces		Double-grooved test-pieces	
		dG/dt (MJ m⁻³)	$\alpha\ (= d/t)$	dG/dt (MJ m⁻³)	$\alpha\ (= d/t)$
Quenched					
25	150	70	0.47	65	0.43
50	220	78	0.35	48	0.22
75	100	90	0.90	62	0.62
100	30	32	1.05	22	0.73
Annealed					
25	40	7	0.18	6	0.15
50	145	6	0.04	5.5	0.04
75	105	–	–	4	0.04
100	32	25	0.78	16	0.50

Figure 5 Sketch of plastic zone cross-section for single-grooved and double-grooved test-pieces (schematic).

at both edges of the tear path and the volume undergoing plastic deformation is correspondingly reduced (Fig. 5b).

Further evidence for attributing the large values of tearing energy for thick test-pieces to plastic deformation in a volume of material whose cross-sectional area is governed by geometrical considerations, is provided by experiments with test-pieces of Type II, shown in Fig. 3. When the height h of the connecting web was small in comparison with the tear path width t, then the fracture energy was found to increase in proportion to t, exactly as for Type I test-pieces (Fig. 6). On the other hand, when the height h of the web was comparable to or larger than the width t, then the measured tear energy was found to be largely independent of t (Fig. 7), and now varied in proportion to h (Fig. 8). The size of the zone undergoing plastic deformation in Type II test-pieces is shown schematically in Fig. 9, to illustrate the change from a proportionality to t^2 to a proportionality to th as the height h increases in comparison to the width t. The plastic work G_p per unit width of tear path thus becomes proportional to t when h is small, and to h when t is small, which accounts for the form of the experimental relations shown in Figs 6 to 8.

Thus, for both Type I and Type II test-pieces the plastic zone is finite in extent and restricted to a region near the tear tip. This is because local stress concentrations associated with sharp edges cause plastic yielding here first. Stresses far away from the tear edges are lower, and do not cause yielding. For ungrooved test-pieces, however, the stress field may be sufficiently homogeneous for the whole cross-section to undergo plastic yielding. Failure by tearing then becomes impossible because plastic deformation of the entire test-piece takes place before rupture.

3.2. Effect of annealing

The tear energy of annealed HDPE was found to be much smaller than for quenched HDPE. It was also less dependent upon the thickness of the test-piece or the width of the tear path, Table II. This is presumably due to the less ductile nature of the annealed material, Table I. Only a small zone around the tear tip appeared to under-
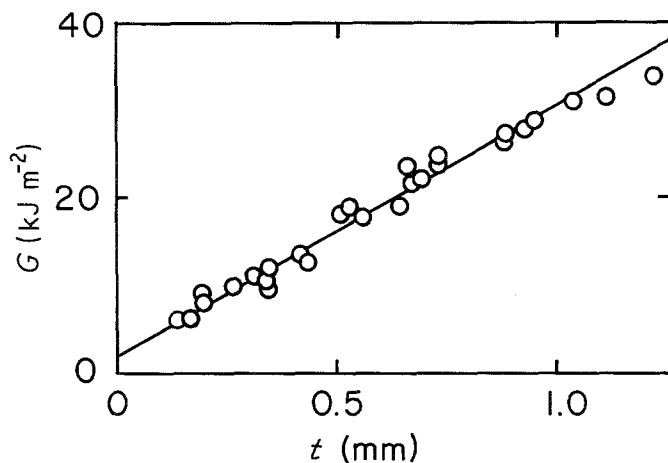

Figure 6 Dependence of tear energy G upon tear path width t for Type II test-pieces of HDPE. $h = 0.1$ mm, tear rate $= 0.4$ mm sec^{-1}, temperature $= 25°$ C.
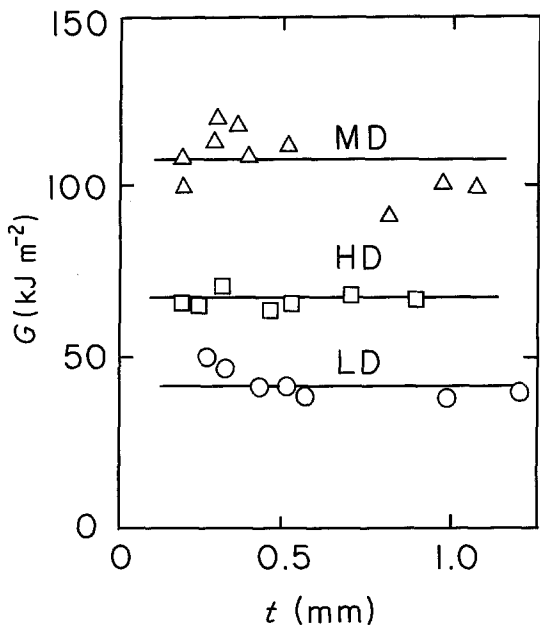
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Figure 7 Dependence of tear energy G upon tear path width t for Type II test-pieces. $h = 0.43$ mm, tear rate = 0.4 mm sec^{-1}, temperature = 25° C.

go plastic yielding and its contribution to the observed tear energy was correspondingly small. It is noteworthy, however, that the extrapolated tear energy G_0 for test-pieces of zero thickness was found to be unaffected by annealing. At all tear rates and test temperatures the values for quenched and annealed test-pieces were

substantially the same. Thus, G_0 appears to be a valid measure of the "intrinsic" tear strength of HDPE, independent of large-scale plastic yielding in the material.

Although generally much smaller than for thick test-pieces the values obtained for G_0 were still relatively large, about 1 to 20 kJ m^{-2}, in comparison with theoretical and experimental estimates of the fracture energy for hydrocarbon elastomers in the absence of dissipation processes, about 20 to 100 J m^{-2} [25–27]. It therefore appears that substantial contributions from dissipative processes are still present. This aspect is discussed further in the following section.

The tear energies of low-density and medium-density polyethylene were not significantly affected by the annealing procedures used in the present experiments: 1 to 1.5 h at temperatures ranging from 100 to 120° C, followed by slow cooling at about 1° C min^{-1}. The values obtained were

$$G_c \text{ (kJ m}^{-2}) = 10 + 35\, t \text{ (mm)}$$

for single-grooved test-pieces of LDPE at room temperature (25° C) and

$$G_c \text{ (kJ m}^{-2}) = 10 + 90\, t \text{ (mm)}$$

for MDPE.

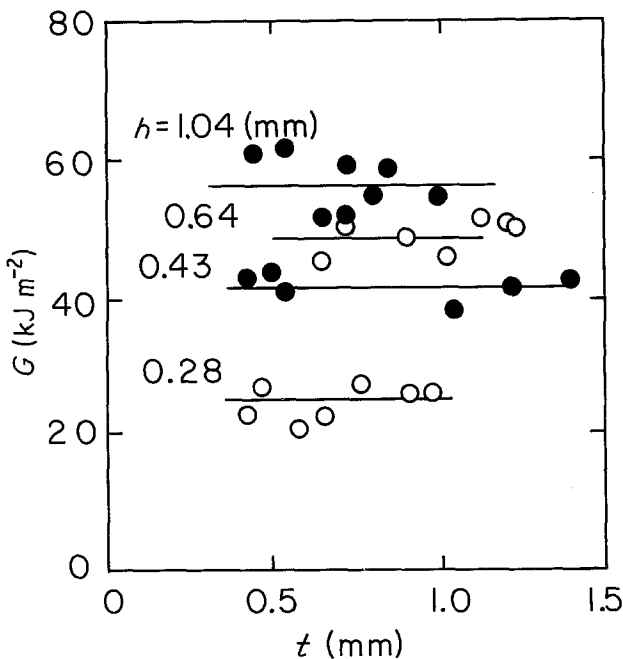*Figure 8* Dependence of tear energy G upon tear path width t and web height h for Type II test-pieces of LDPE. Tear rate = 0.4 mm sec^{-1}, temperature = 25° C.
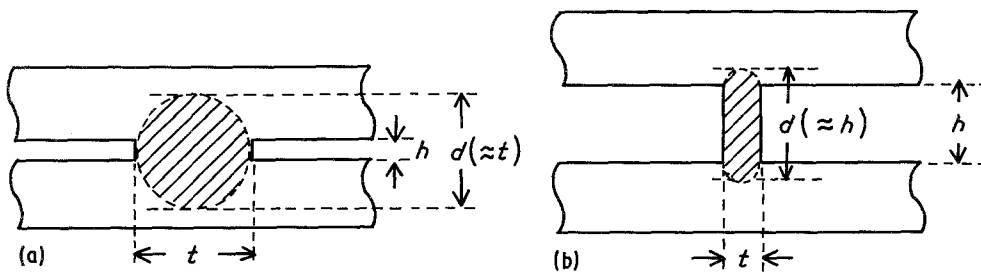
Figure 9 Sketch of plastic zone cross-section for Type II test-pieces (schematic).

3.3. Effect of rate of tearing and temperature

Values of the fracture energy G_0 for test-pieces of zero thickness (obtained by extrapolation) and G_m for test-pieces about 0.6 mm thick are plotted against the logarithm of the rate R of tear propagation in Figs. 11 and 12. In each case, experimental relations are also shown for several temperatures.

Although the results for thin and thick test-pieces differ greatly in absolute magnitude, and somewhat in general form, they have several features in common. In both cases the fracture energy varies strongly with tear rate, passing through a maximum at a particular rate of tearing denoted R_1. This characteristic rate was found to be substantially the same for both G_0 and G_m, but was itself a strong function of tem-perature, increasing by a factor of about 3000 as the temperature was raised from 25 to 100° C. Values of R_1 are given in Table III.

This variation in tear energy with rate of tearing is strikingly similar to the variation found in the dissipative properties of polyethylene with frequency of small oscillatory deformations, termed the α-absorption peak. In Fig. 13, experimental measurements by Takayanagi [28], of the loss modulus E'' for HDPE are plotted against the angular frequency ω of mechanical oscillation. These relations are also seen to pass through maximum values at characteristic frequencies, denoted here ω_1, which are in turn a strong function of temperature, increasing by orders of magnitude as the temperature is increased from 25 to 100° C.

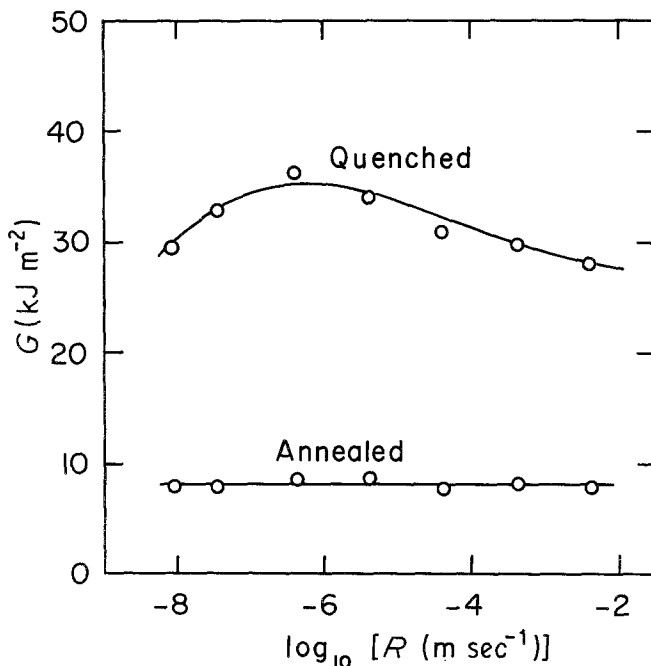Indeed, although the E'' relations differ

Figure 10 Dependence of tear energy G upon the rate R of tear propagation for quenched and annealed samples of HDPE.
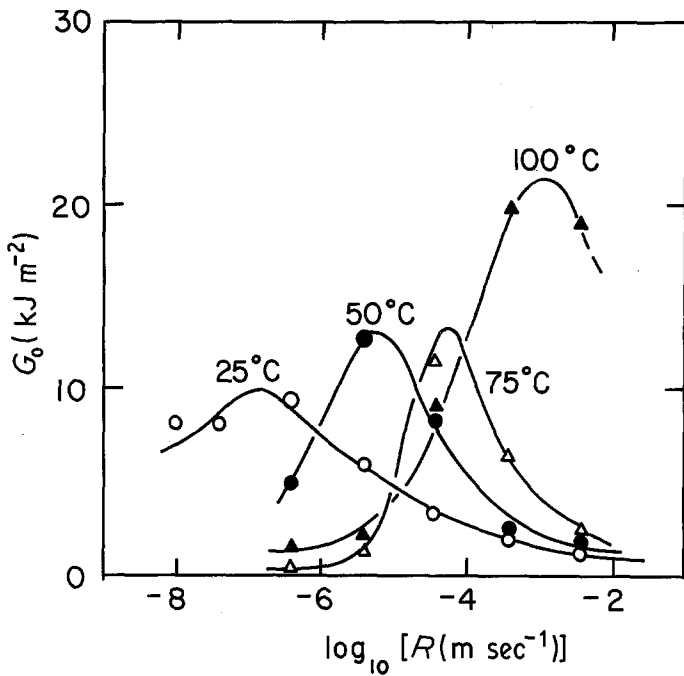
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Figure 11 Dependence of tear energy G_0 ($t = 0$) of HDPE upon the rate R of tear propagation.

significantly in shape from the G_0 and G_m relations shown in Fig. 11 and 12, becoming broader and more diffuse at higher temperatures, there is a close correspondence between the frequencies ω_1 at which E'' passes through maximum values and the corresponding rates R_1 at which the tear energy passes through maxima. Values of ω_1 taken from Fig. 13 are given in Table III for direct comparison.

A general parallel between energy dissipation processes and fracture energy has been widely recognized to hold for elastomers and plastics [13], so that the present correlation between E'' and G is not unexpected. However, in the present case, quantitative comparison can be made between the characteristic rates R_1 and frequencies ω_1. The ratio $\delta = R_1/\omega_1$ presumably denotes a characteristic dimension for the dissipation process at the tear tip. As shown in Table III, this dimension ranges from 1 to $10\,\mu m$,
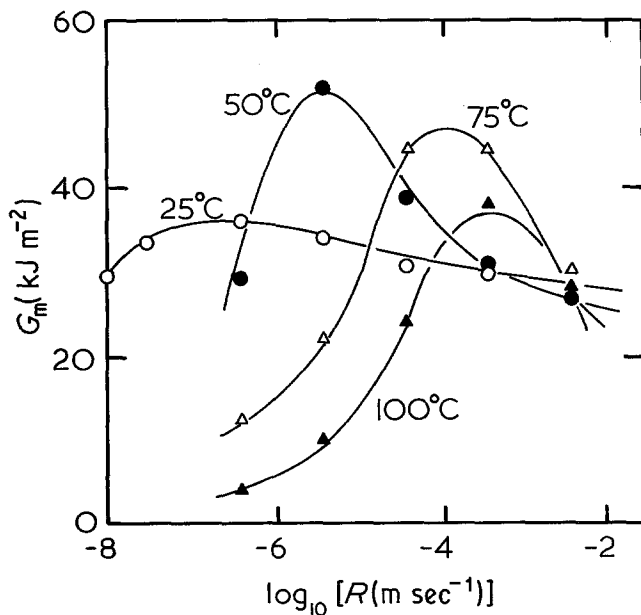

Figure 12 Dependence of tear energy G_m ($t = 0.6$ mm) of HDPE upon the rate R of tear propagation.

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TABLE III Comparison between rates R_1 at which the tear energy is a maximum and angular frequencies ω_1 at which E'' is a maximum

Temperature ($^\circ$C)	$\log_{10} R_1$ (m sec^{-1})	$\log_{10} \omega_1$ (rad sec^{-1})	δ ($= R_1/\omega_1$) (μm)
25	-6.75	-1.2	2
50	-5.4	-0.4	10
75	-4.1	$+3.1$	4
100	-3.1	$+3.0$	1

averaging about $4\,\mu$m, over the entire range of temperatures. Apparently the dissipation process takes place on a scale of about $4\,\mu$m during tearing.

This dimension is strikingly similar to the reported spherulite size for HDPE [1] and suggests that the tear process involves energy dissipation by spherulite deformation in the same way that the α-absorption peak does. This mechanism is complex [28] and still somewhat uncertain, but the present correlation suggests that, whatever its nature, it is the principal cause of energy dissipation during tearing.

From studies of mechanical fatigue cracking, the size of intrinsic flaws in LDPE has been deduced to be about the same as the spherulite diameter in that material: between 20 and 60 μm

[29]. Apparently, inter-spherulite boundary cracks can nucleate failure by mechanical fatigue. The present observations are rather difficult. They suggest that crack growth involves extensive deformation of material in a layer about one spherulite diameter in width. Thus, large-diameter spherulites would be preferred on the latter basis because the effective width of the tear path would be greater and, hence, the energy required for fracture would be greater also, whereas small-diameter spherulites would be preferred on the former basis, because the effective size of intrinsic flaws would be correspondingly small. However, the amount of energy expended in local deformation is not necessarily proportional to the effective width of the tear path, even when that is prescribed by the size of the spherulites present. A more detailed study of the effect of spherulite size and structure upon the magnitude of the fracture energy is clearly needed.

4. Conclusions
The following conclusions are obtained.

1. The tear energy of polyethylene and, presumably, other ductile materials, depends upon the volume of material that yields before the tear
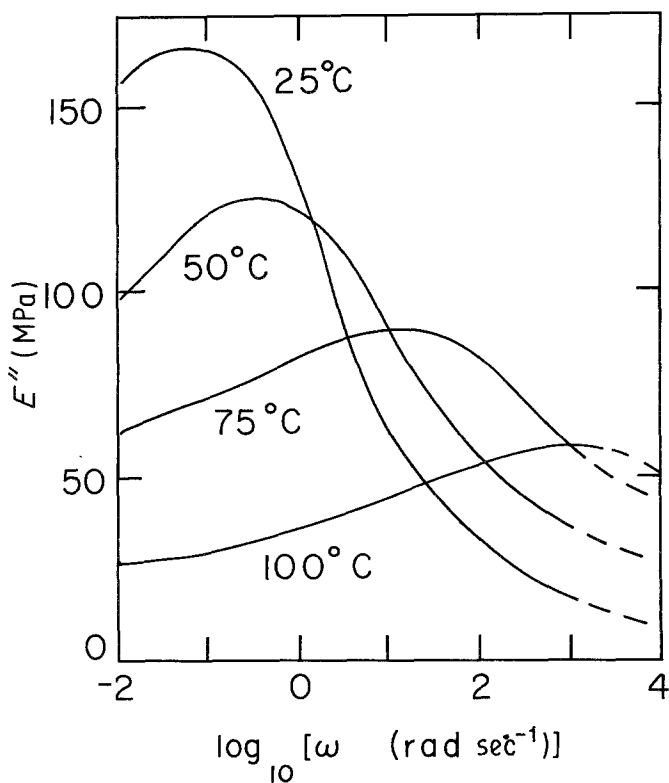
Figure 13 Loss modulus E'' of HDPE against angular frequency ω of mechanical oscillation. (Data taken from [28]).

propagates. The work of local plastic deformation is then included in the tear energy. In consequence, the tear energy of films increases with the film thickness, or, for Type II test-pieces, with the height of the connecting web.

2. By extrapolation, a threshold value of the tear energy can be deduced for a film of zero thickness, i.e. in the absence of local plastic deformation. The values obtained are still relatively large, however: 1 to $20 \, \text{kJ m}^{-2}$ for HDPE.

3. The tear energy for HDPE varies strongly with the rate of tear propagation, passing through a maximum at a characteristic rate R_1. This rate is itself a strong function of temperature, increasing by a factor of about 3000 as the temperature is raised from 25 to 100° C.

4. By comparing the rate dependence of tear energy with the reported variation of the loss modulus E'' with frequency for HDPE, a characteristic dimension δ is obtained for the tear process of $4 \, \mu\text{m}$, strikingly similar to reported spherulite sizes for HDPE. It is concluded that dissipation of mechanical energy in a region of the material having a thickness of this magnitude is largely responsible for the observed tear energy in thin test-pieces, and, together with local yielding processes, in thicker ones.

5. On annealing HDPE, the plastic yielding process is suppressed and the tear energy falls from values of about $50 \, \text{kJ m}^{-2}$ to a value close to threshold values. No comparable decrease was found on annealing MD or LD polyethylene.

Acknowledgements

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