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# The initiation of slow crack growth in linear polyethylene under single edge notch tension and plane strain

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The kinetics and microstructural changes associated with the initiation of slow crack growth in PE were measured. The initiation process consists of an instantaneous deformation zone which grows at a constant velocity until the beginning of fracture. The velocity of the damaged zone accelerates when the fibril fracture begins at the root of the initial notch. It was found that the initial velocity of the deformation zone depended on stress to about the 4th power and had an activation energy of about 100 kJ mol$^{-1}$; these results are about the same as those found by Chan and Williams for the crack growth velocity. It is concluded that both crack initiation and crack growth are governed by the same fundamental process, notably fibril thinning.

## 1. Introduction

Under a low stress and after a long time, polyethylene exhibits creep fracture which appears brittle on a macroscopic scale. The crack growth is preceded by an incubation period which involves the formation of a coarse type of craze. Since the time to incubate crack growth is a significant part of the total time to failure, it is important to understand the kinetics of the initial deformation zone and its relationship to the subsequent growth of the crack.

We have examined [1] the microstructure of the deformation and fractured zone that emanates from a notch in PE which was exposed to plane strain. The results are depicted in Fig. 1 where the leading edge of the damaged zone consists of crack-like pores followed by a region of increasing fibrillation and finally by a region of fibril fracture. Since the fibril diameter and thickness of the deformation zone tend to be at least an order of magnitude larger than those observed in most polymers, including PE at low temperatures, the deformation zone may be considered to be a coarse type of craze.

We also examined [1] the growth of the deformation zone and found that its dimensions increased linearly with time under a constant stress. The size of the damaged zone that occurs instantly upon loading was measured as a function of stress and depth of the initial notch. The size was approximately that predicted by the Dugdale model. It was also found that above one-half of the yield point the deformation zone was balloon-like in shape and planar below.

In this paper the incubation of crack growth was examined further. The transition from the growth of the initial deformation zone to the beginning of crack growth was observed from the kinetic and the macroscopic viewpoints. All tests were under plane strain conditions as usually occurs in engineering structures.

## 2. Experimental details

The polyethylene was the same Marlex 6006

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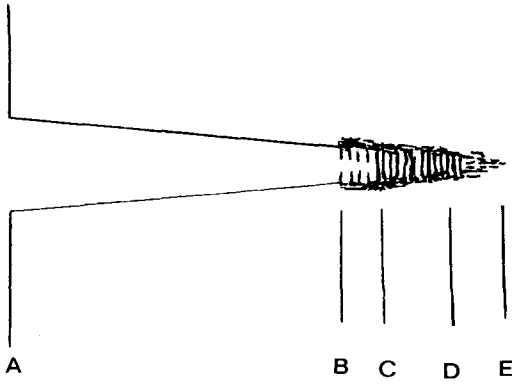


*Figure 1* Schematic illustration of damaged zone: AB notch, BC fractured, CD fibrillation, DE microcracks.

supplied by Phillips Petroleum as in the previous investigations: $M_n = 19\,600$; $M_w = 130\,000$, density = $0.964\,g\,cm^{-3}$, yield point = 25 MPa at 296 K and strain rate of $0.02\,min^{-1}$. The material was compression moulded at Phillips into 4 mm thick plaques according to ASTM D1928 (cooling rate = $15°\,C\,min^{-1}$).

The notched tensile specimens (Fig. 2) were 4 mm thick and 18 mm wide with a single surface notch running along the width of the specimens. The average depth of all notches was 0.25 mm. Previous experiments [1] showed that about 96% of the damaged zone was exposed to plane strain conditions. The notches were made with a razor blade under very well controlled conditions so that no visible damage was observed at the root of the notch prior to loading. The details of notching are in a previous paper [2].

The damaged zone at the root of the notch was observed by several microscopic techniques: (1) optical transmission of thin slices from the middle of the specimen (Fig. 3); (2) scanning electron microscope (SEM) observations of the plane of damage after exposure by rapid fracture in liquid nitrogen (Fig. 4); and (3) the surface shown in Fig. 3 was etched with permanganic acid according to the method of Bassett and Hodge [3] and modified by Naylor and Phillips [4] and then observed with the SEM as in Fig. 5. Generally the length of the damaged zone as exhibited by Fig. 3 was slightly greater than the sharp boundary of Fig. 4. Since the leading edge of the deformation zone consisted of an array of crack-like pores, it was more convenient to measure the velocity of the deformation zone from the sharp boundary such as in Fig. 4. Most measurements of the length of the damaged zone were made at a magnification of $\times 160$ and the uncertainty in the length of the sharp boundary shown in Fig. 4 was about $\pm 0.005$ mm.

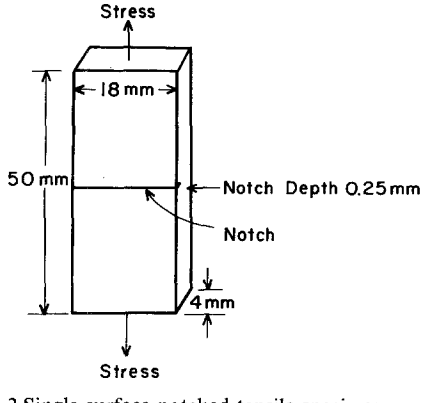


*Figure 2* Single surface notched tensile specimen.

The value of stress intensity, $K$, was calculated from the following equation [5]:

$$K = 1.12\pi^{1/2}\sigma a_0^{1/2} \quad (1)$$

where $\sigma$ is the applied stress and $a_0$ is the notch depth. The uncertainty in $K$ was about $\pm 4\%$.

In these experiments the major variables were the stress, time and the temperature. The stresses were kept below one-half the yield point so that the zone of damage would be planar rather than balloon-like as shown in the previous paper [1].

## 3. Results

Fig. 6 shows the length of the damaged zone plotted against time. The damage zone was measured from micrographs such as shown in Figs. 3 and 4. In Fig. 6 the notch depth and the applied stress were constant and the curves for 25, 30, and 40° C were obtained. All curves such as those in Fig. 6 showed (1) an instantaneous deformation at zero time, (2) a linear region, and (3) and an accelerating region. In Fig. 6, the 25° C curve was not extended far enough to include the accelerating region. The length of the damaged zone at the end of the linear range increased with decreasing temperature.

Optical micrographs that correspond to various times for the 30° C curve in Fig. 6 are shown in Figs. 7a to e. These optical micrographs were obtained on thin slices which were taken from the centre of the damaged specimen and then were slowly loaded to a fixed stress under the optical microscope. The purpose of the subsequent loading was to open the notch in order

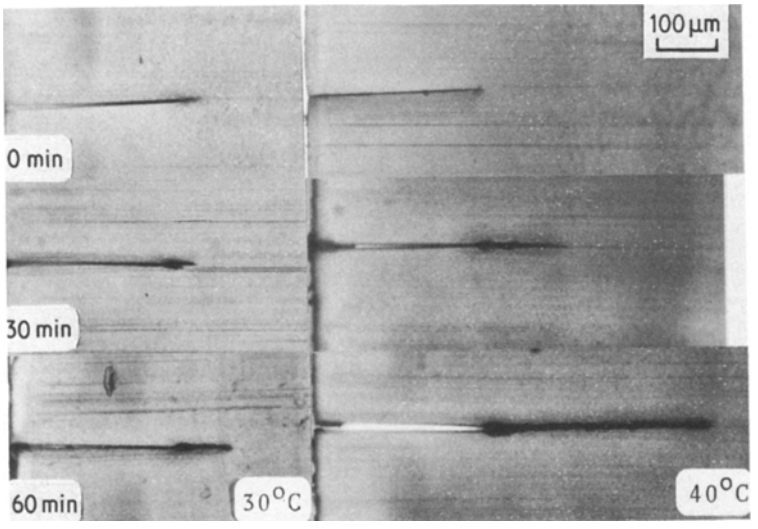


*Figure 3* Transmission optical micrographs of damaged zone. Specimen about 0.2 mm thick.

to reproduce the state of the specimen when it was under load. Figs. 7a to e show that when the period of acceleration begins, fibril fracture commences. Fig. 7f which is a higher resolution micrograph of Fig. 7c shows that fracture began at 200 min where the linear region ends.

The log of velocity of the deformation zone in the initial linear region is plotted against log stress in Fig. 8 for 30° C and $a_0 = 0.25$ mm. The resulting relationship is

$$V = 6.3 \times 10^{-8} \sigma^{4.2} \text{ mm min}^{-1} \quad (2)$$

The velocity of the deformation zone in the linear region from Fig. 6 was plotted against $1/T$ as shown in Fig. 9. From the slope of the curve, an activation energy of $97 \text{ kJ mol}^{-1}$ was obtained.

The morphology of the deformation zone was examined with the SEM at $\times 2500$ after fracture in liquid nitrogen (Fig. 10). The corresponding optical micrograph is Fig. 7e where the location

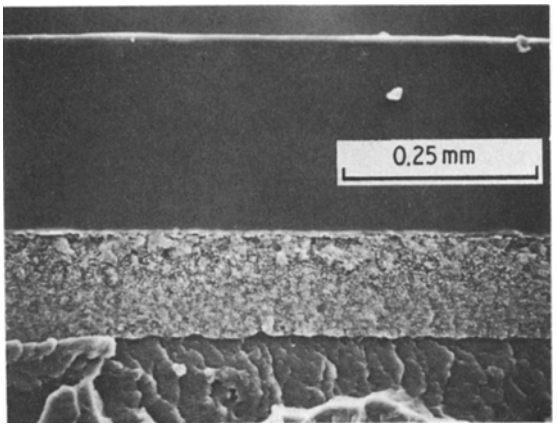


*Figure 4* Scanning electron micrograph of specimen after fracture in liquid nitrogen. Tested at 30° C, 10 MPa, 90 min. Same as Fig. 7b.

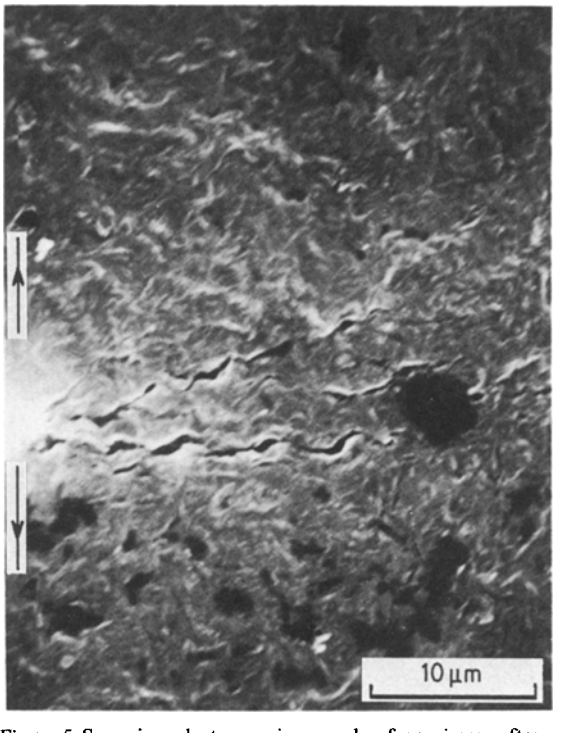


*Figure 5* Scanning electron micrograph of specimen after etching in permanganic acid. Tested at 30° C, 10 MPa, 30 min. Same as Fig. 7a. Arrows show stress.

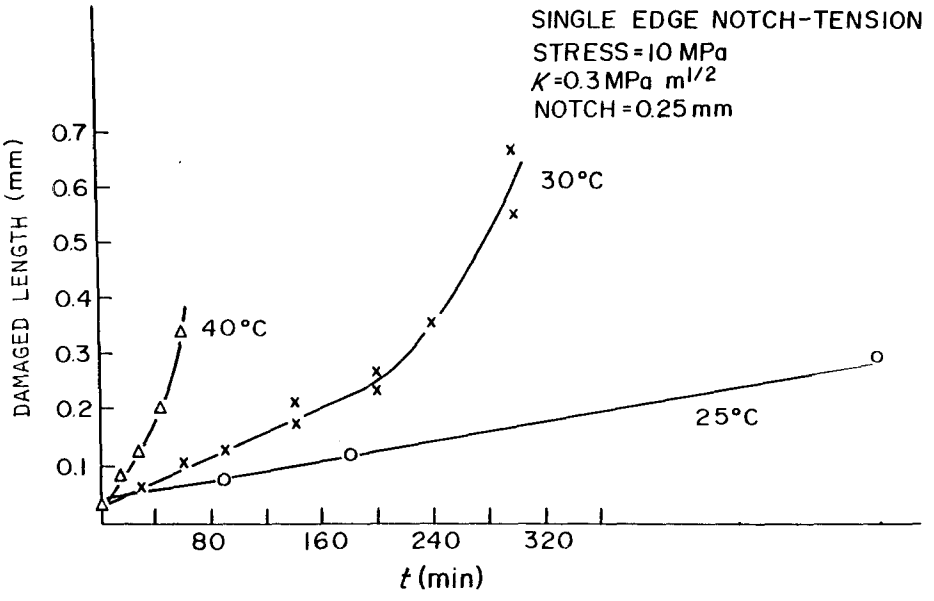


*Figure 6* Length of damaged zone plotted against time at 25.5, 30 and 40° C.

of each field of view in Fig. 10 is designated. The morphology is most coarse and constant in the region where fracture had occurred. Then the morphology becomes finer and finer until the undamaged region is reached. This increase in coarseness of the morphology from the tip toward the fractured region most likely corresponds to an increase in the porosity of the craze. The increase in porosity is caused by the increase in the interfibrillar spacing as the damage progresses toward the fractured region. These observations in addition to the bifurcation of the damage at the tip of the damage zone (Fig. 5) are consistent with the schematic illustration of the damaged zone illustrated in Fig. 1.

## 4. Discussion

Fig. 6 shows the typical kinetics for the initiation of crack growth. There is an initial deformation which occurs instantly upon loading the specimen and which is small compared to the size of the initial notch. The deformation zone grows at a constant velocity. The deformation zone begins to accelerate when fibril fracture occurs which represents the beginning of crack growth.

It is expected that the time independent Dugdale model [6] can be used as a basis for predicting the length of the instantaneous deformation zone, $\Delta a_0$ where

$$\Delta a_0 = \frac{\pi}{8} K^2/\sigma_y^2 \tag{3}$$

where $K$ is the stress intensity and $\sigma_y$ is the yield point. For $\sigma_y = 25\,\text{MPa}$ and $K = 0.316\,\text{N m}^{-3/2}$, $\Delta a_0 = 0.063\,\text{mm}$. Part of the experimental value for the instantaneous deformation is obtained from Fig. 6 at $t = 0$. However, Fig. 6 is based on the sharp boundary observed in a micrograph such as Fig. 4, but the deformation zone extends about 0.03 mm beyond this sharp boundary as determined by corresponding measurements using the optical micrograph such as Figs. 3 and 7. Thus, the experimental value of the instantaneous deformation is 0.055 mm for 30° C and $K = 0.316$, which agrees nicely with the value of 0.063 mm from the Dugdale model.

The fact that the velocity of the crazed deformation zone is constant prior to crack growth is not expected. Many polymers that craze in air exhibit a decreasing velocity with time. Kinloch [7] explained the time dependent velocity by modifying the Dugdale model. He assumed that $\sigma_y$ decreased with time in accordance with the equation

$$\sigma_y = \varepsilon_y E_0/t^m \tag{4}$$

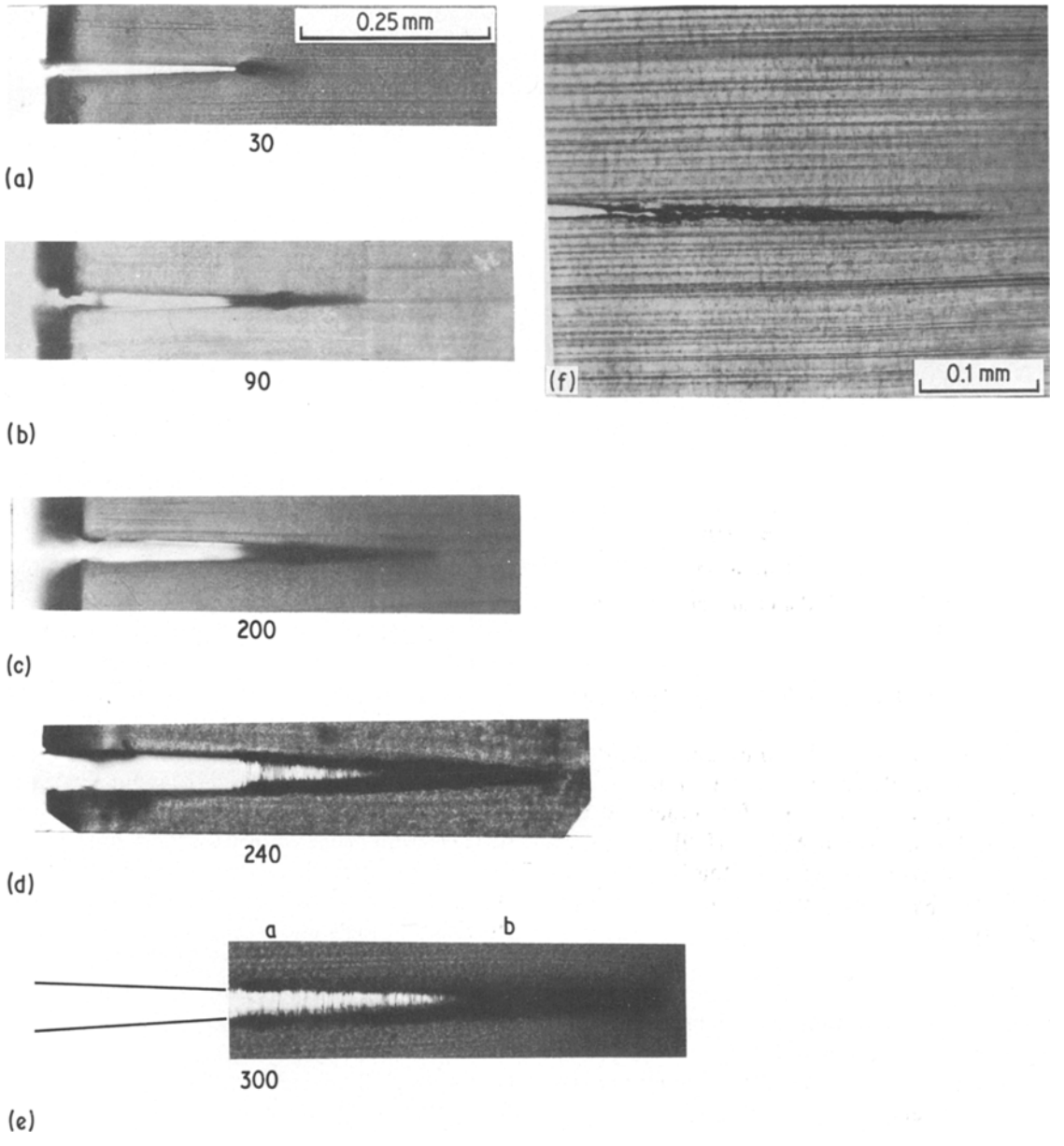


*Figure 7* Optical micrographs for specimens exposed to 10 MPa and 30° C corresponding to the various times for curve in Fig. 6. The thin films were photographed under stress to expose the fibrilla structure. (a) 30 min, (b) 90 min, (c) 200 min, (d) 240 min, (e) 300 min. (f) Same as (c) but thinner slice, 0.02 mm thick, gives higher resolution.

where $\varepsilon_y$, the yield strain, was assumed to be constant and $E_0$ is the unrelaxed modulus. Thus, the velocity of the deformation zone

$$V = \pi/8 \; K^2 t^m / \varepsilon_y E_0 \qquad (5)$$

Measurements [7] of craze velocity against time give values for $m$ of 0.113, 0.075, 0.04 for PMMA, rubber modified PS, and PC, respectively. Sauer and Hsiao [8] and Argon and Salama [9] observed a constant craze velocity in PS. These differences in behaviour, including our observation with PE, are not understood at this time.

That the deformation zone begins to accelerate with crack growth is most obviously influenced by the increase in stress intensity as the initial notch lengthens. The question arises as to whether the velocity of the initial

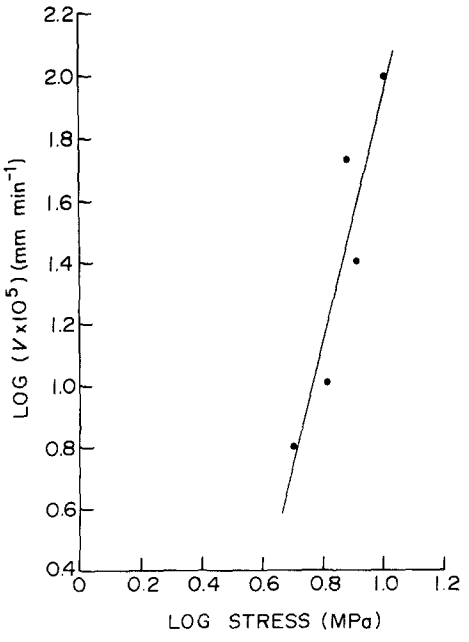


*Figure 8* Log of velocity of deformation zone plotted against log stress at 30° C.

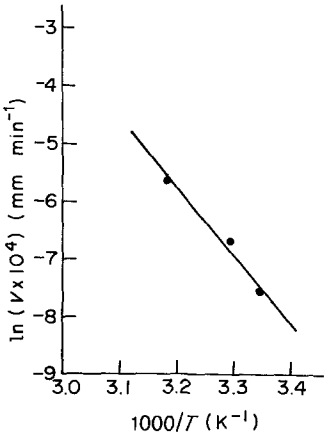


*Figure 9* Natural log of velocity plotted against $1/T$.

deformation zone is related to the crack growth velocity.

The velocity of the deformation zone prior to crack growth described by Equation 2 is very similar to the equation for crack growth observed by Chan and Williams [10] on the same type of polyethylene as used in this investigation. The work by Chan and Williams [10] on crack velocity gave

$$V = A\sigma^4, \tag{6}$$

where $A$ is a constant that depends on temperature and geometry. The difference in exponents between 4 and 4.2 is within the experimental scatter.

Chan and Williams [10] also found an activation energy of 105 kJ mol$^{-1}$ for the crack velocity which is nearly equal to our value of 97 kJ mol$^{-1}$. The agreement between these data indicates that the same basic mechanism governs the initiation of crack growth and crack growth itself.

All the microscopic observations show that the development of fibrillation is the underlying microcopic process. In order for the deformation zone to increase in length prior to crack growth, it is necessary for the deformation zone to widen. This widening process is controlled by the rate of fibril extension. In order for the crack to grow, the extension of the fibrils is required and then the fibrils break when they reach a critical cross-sectional area. Since the crack growth rate and the velocity of the deformation zone both have the same dependence on stress and the same activation energy, it appears that the rate of fibril extension is the rate determining process rather than the rate of final fracture of the fibrils. The process of fibril fracture was observed in a previous paper [2] and the observations agree with the above viewpoint since the fibrils spend most of their life extending and the final fracture occurs in a very short time.

It is also interesting to note that Wilding and Ward [11] measured the creep rate of oriented PE fibres of the same type of PE as ours. They observed an activation energy of 120 kJ mol$^{-1}$ which is close to the values for deformation zone velocity and crack growth, considering that the fibrils in the deformation zone of a notch may not be as well oriented as the highly oriented fibres of Wilding and Ward. It is therefore concluded that the thinning of the fibrils is the basic process that governs the low-stress long-time fracture in linear PE.

The submicroscopic mechanisms that control the creep rate are, however, not known. One mechanism is simple chain sliding and the other is chain rupture. The activation energy of about 100 kJ mol$^{-1}$ is comparable to the $\alpha$ process in polyethylene so that it is now suggested that chain sliding in the oriented fibrils is the fundamental mechanism that governs slow crack growth in PE.

The critical amount of fibril creep that is required for fibril fracture and the initiation of

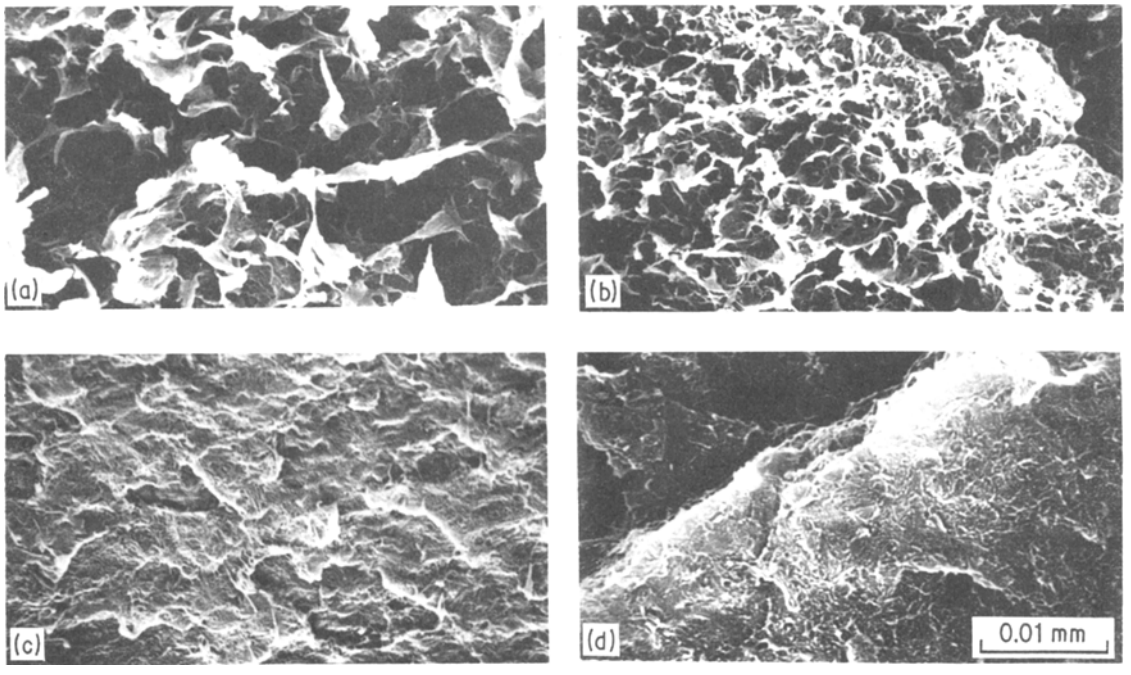


*Figure 10* Morphologies of damaged zone seen by SEM after fracture in liquid nitrogen. Specimen exposed to 10 MPa at 30° C for 300 min. Fig. 7e is the corresponding optical micrograph. (a) Fractured region, (b) fibrillated region, (c) tip of deformation zone, (d) virgin material.

crack growth is not known. Many theories of microfracture mechanics have proposed that a critical crack opening displacement determines crack growth. In the case of crazing or fibrillation, the concept of a critical crack opening displacement represents a simplified viewpoint because two processes contribute to the widening of the deformation zone. As observed by Bhattacharya and Brown [2], the fibrils lengthen by thinning the existing fibril and at the same time additional fibrilation forms by end-growth from the edge of the deformation zone. Kramer [12] has also observed the same behaviour with respect to crazing in general. Work by Döll [13] and Trassaert and Schirrer [14] also support this viewpoint. Probably the critical condition for the initiation of crack growth depends on a critical amount of thinning of the fibril and its degree of orientation strengthening. Carr *et al.* [15] have emphasized the importance of orientation strengthening in determining the fracture behaviour of PE.

## 5. Conclusions

1. The initiation of crack growth in prenotched PE involves an instantaneous deformation followed by a period of constant velocity for the deformation zone.

2. The velocity of the damaged zone begins to accelerate when fibrils at the root of the notch begin to fracture.

3. The length of the deformation zone produced during the loading of the specimen is predicted by the Dugdale model.

4. The tip of the deformation zone consists of an array of microcracks which then transform to an increasingly fibrillated structure toward the root of the notch.

5. The temperature dependence of the initial velocity of the deformation zone is about $100\,\mathrm{kJ\,mol^{-1}}$.

6. The initial velocity of the deformation zone is related to stress by $V = A\sigma^n$ where $n \approx 4$.

7. The agreement between the values of $n$ and activation energy from measurements of crack growth velocity and the velocity of the initial deformation zone indicate that both are controlled by the same mechanism.

8. It is proposed that thinning of the fibril by creep is the underlying mechaism that controls both crack growth and the initiation time for crack growth.

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## References

1. S. K. BHATTACHARYA and N. BROWN, *J. Mater. Sci.* **20** (1985) 2767.
2. *Idem, ibid.* **19** (1984) 2519.
3. D. C. BASSETT and A. M. HODGE, *Proc. Roy. Soc. London* **A377** (1981) 25.
4. K. L. NAYLOR and P. J. PHILLIPS, *J. Polym. Sci.* **21** (1983) 2011.
5. P. C. PARIS and C. G. SIH, Proceedings ASTM Technical Publication No. 381 American Society for Testing and Materials, Philadelphia (1965) p. 30.
6. J. K. KNOTT, "Fundamentals of Fracture Mechanics" (Butterworths, London, Boston, 1973) Ch. 3.
7. A. J. KINLOCH, *Met. Sci.* **14** (1980) 305.
8. J. A. SAUER and C. C. HSIAO, *ASME Trans.* **75** (1953) 895.
9. A. S. ARGON and M. M. SALAMA, *Phil. Mag.* **36** (1977) 1217.
10. M. K. V. CHAN and J. G. WILLIAMS, *Polymer* **24** (1983) 234.
11. M. A. WILDING and I. M. WARD, *ibid.* **22** (1981) 870.
12. E. J. KRAMER, "Crazing in Polymers", Advances in Polymer Science 52/53, edited by H. H. Kausch (Springer Verlag, Berlin, New York, 1983) p. 14.
13. W. DÖLL, *ibid.*, p. 105.
14. P. TRASSAERT and R. SCHIRRER, *J. Mater. Sci.* **18** (1983) 3004.
15. S. H. CARR, B. CRIST and T. J. MARKS, "Structural Basis for the Mechanical Properties of Polyethylene", Final Report to Gas Research Institute, Chicago (1984).

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