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The transition from ductile to slow crack growth failure in a copolymer of polyethylene

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The failures of ethylene–hexene copolymer single-edge notch tensile specimens were observed under a constant tensile load. The notch opening was measured against time over a range of stress. Three failure modes were observed: ductile, brittle and transitional. The microscopic changes at the notch tip were correlated with each of the modes of failure. Early in the test the ultimate mode of failure can be predicted from the microstructural changes in the notch. In the transition region, the lifetime increases as the stress increases because the blunting of the notch offsets the effect of the applied stress–stress field. The ductile failure is controlled by the macroscopic creep behaviour and the brittle failure occurs by slow crack growth that starts at a craze.

1. Introduction

In the neighbourhood of room temperature and above, PE undergoes a ductile–brittle transition. The ductile mode occurs by a creep process under stresses greater than about one half the yield point and for stresses less than one half the yield point, failure occurs by slow crack growth. The brittle mode is associated with fracture under plane strain conditions and as in all brittle fracture processes, the crack is initiated at a pre-existing point of stress concentration such as a sharp notch.

It is the purpose of this paper to correlate the changes in the microscopic structure at the root of a notch with the kinetics of the ductile and brittle modes of failure. Brown, Donofrio and Lu [1] formulated a quantitative description of this ductile–brittle transition. It was shown that the time to failure by the ductile mode depended on the bulk creep behaviour as it is affected by stress and temperature. The time to failure by the brittle mode depended on the nucleation and growth of a crack as it depended on stress, temperature and the depth of the pre-existing notch. Each process has its own kinetics and both damage processes occur simultaneously with high stress favouring the ductile process and low stress the brittle process. The mode of final failure depends on which process is faster for a given stress, temperature and notch depth [2–5]. It is shown in this investigation that microscopic changes at the root of the initial notch nicely reflect the competition between the two modes of failure and that microscopic observations of the root of the notch give an early indication of whether the ultimate failure will be by the ductile or brittle mode.

2. Experimental

The PE used in this investigation is the same resin that is used for gas pipes. It is one of the toughest types of PE for resisting slow crack growth which is the important mode of failure in gas piping systems. The material is an ethylene–hexene copolymer with 4.5 butyl chains per 1000C; density = 0.936, $\bar{M}_n$ = 15 000 and $\bar{M}_w$ = 170 000. The specimens were machined from 10 mm thick compression moulded plaques which were very slowly cooled from 190° C. The geometry of the test specimens is shown in Fig. 1 where the width of the specimen was wide enough to ensure that over 90% of the fracture process was under plain strain conditions and the notch depth was optimized to minimize the time to failure while ensuring that the fracture mode was brittle. The side grooves shorten the time to failure and reduce the amount of plane stress fracture that occurs at the edge of the crack. Fig. 2 shows the method for observing the crack growth process via the notch openings AA at the surface, CC at the root of the notch as formed by a razor blade and BB at the base of the craze which is nucleated by the notch. The constant stress was applied by means of a 5 : 1 to lever ratio. The notch was produced by pressing a fresh razor blade into the specimen at a speed of 50 μm min^{-1}.

3. Results

The effect of stress and temperature on the time to failure is shown in Fig. 3. In the ductile region the time to failure depends on stress as $t_f \sim \sigma^{-34}$ and in the brittle region as $t_f \sim \sigma^{-3.3}$. For a given stress, t_f exponentially increases as the temperature decreases. In the transition zone between the ductile and brittle regions there is a knee in the curves which is anomalous in the sense that t_f decreases as σ decreases.

Fig. 4 shows typical kinetics for the brittle region where the notch openings AA, CC and BB are plotted

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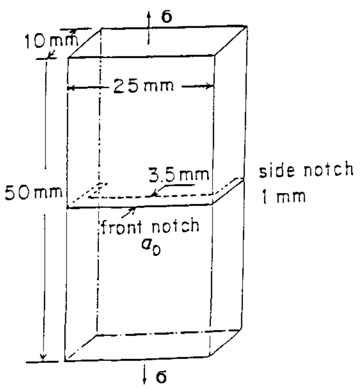

Figure 1 Geometry of specimen.

against time. The initial value of the notch opening occurs immediately upon loading the specimen. It is important to note that the rate of notch opening is small prior to the initiation of fracture which is indicated by the letter c on the BB curve in Fig. 4. From c to g slow crack growth occurs and after g the remaining ligament yields prior to ultimate failure.

Fig. 5 shows the microscopic changes that occur at the root of the notch for the various times. Fig. 5a shows the damage zone immediately upon loading. The size of the initial damage zone is approximately that predicted by the Dugdale theory; but the aspect ratio of the damage zone, the length divided by the thickness at its base, is somewhat smaller than the Dugdale prediction. Prior to the initiation of fracture, Figs 5a to 5d, the damage zone grows slowly and the aspect ratio remains fairly constant and equal to about 3. In Fig. 5e fracture has initiated at the base of the damage zone and in 5f there is the well developed crack with its process zone.

In Fig. 4, the difference between the CC and BB curves is small compared to the difference between CC and AA curves. This occurs because the amount of creep strain at the root of the notch in the region between CC and BB is small (Fig. 5c). CC − BB ≪ AA − CC is the primary characteristic of the microstructure which indicates brittle behaviour.

The kinetics of the ductile region for notch opening against time are shown in Fig. 6. There is a large difference between the CC and BB curves as compared to the separation between CC and AA. The changes in microstructure corresponding to the times a to f designated on curve BB are shown in Fig. 7. The large amount of creep widens the notch relative to the thickening of the crazed damaged zone. The notch becomes so blunt that its effect as a concentrator of the stress is practically nil. Since the rate of growth of the craze is very dependent on the stress field near the notch, the damage zone hardly grows and the rate of crack growth is suppressed. Thus, the ultimate failure occurs by the yielding of the ligament. At very short times (Fig. 6a) the damage zone is the same shape as in the brittle region because it forms while the notch is still sharp and before it becomes blunt by the creep process.

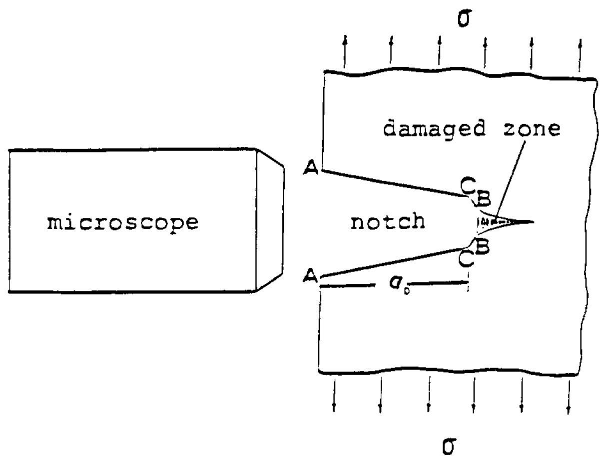

Figure 2 Experimental measurement of the notch opening.

The kinetics of the transition region are shown in Fig. 8. Here the separation between CC and BB is intermediate with respect to the values in the ductile and brittle regions. The corresponding microstructural changes with time are shown in Fig. 9. For short times a to c the damage zone is like that for the brittle region because the notch has not blunted excessively. Finally, the blunting of the notch by creep reduces the growth rate of the damage zone.

The rate of slow crack growth depends on the local stress which is approximately given by the equation

$$\sigma_{\text{local}} = \sigma\left[1 + 2\left(\frac{a}{r}\right)^{1/2}\right] \quad (1)$$

where σ is the applied stress, a the depth of the notch, and r the radius of curvature of the notch. Equation 1 is exact for an elliptic notch. When the applied stress produces brittle fracture, r is small and changes only slightly because the creep rate is very low. As σ increases r increases, but as long as σ_{local}

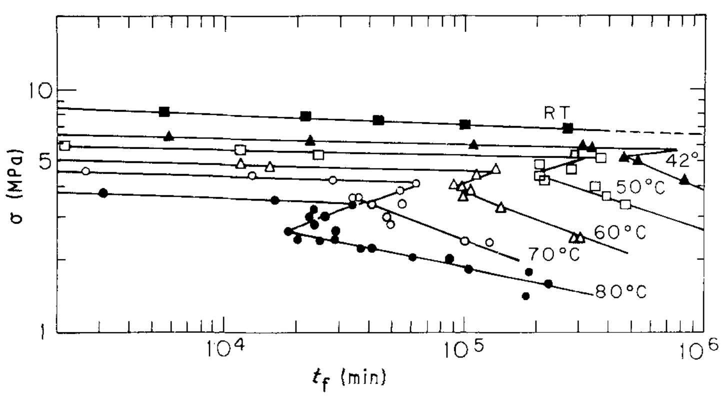

Figure 3 Stress plotted against time to failure (t_f) at various temperatures for copolymer polyethylene with a notch of 3.5 mm. (●) 80° C, (○) 70° C, (△) 60° C, (□) 50° C, (▲) 42° C, and (■) room temperature.

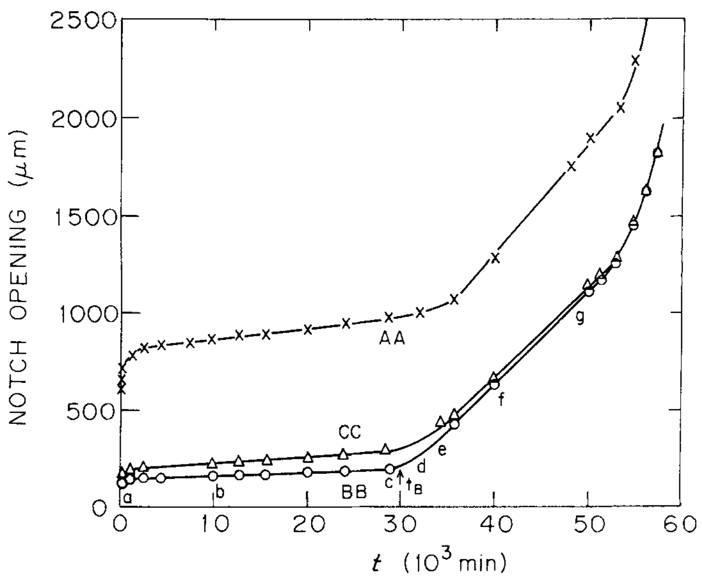

Figure 4 Notch openings AA, CC, and BB plotted against time in brittle region of failure, tested at 80° C and 2.0 MPa. The arrow indicates the beginning of slow crack growth.

increases the crack growth rate increases and time for brittle failure decreases.

A further increase of σ into the transition region increases r to such an extent that the local stress is decreased and the time to failure by crack growth is increased. Finally, when σ is further increased into the ductile region r becomes so large that the stress concentration of the notch becomes very small and at the same time the creep rate increases so that the ligament yields before appreciable crack growth occurs.

The question arises as to how the presence of the notch affects the kinetics of ductile failure. Fig. 10 shows stress against time to failure for a specimen with a 3.5 mm notch and the ordinary dumbbell specimen without a notch. The dependence of time to failure on stress, in the case of the notched specimen the stress is based on the ligament area. The curves of σ against time to failure are practically the same for the notched and unnotched specimens. The slight shift between the curves comes from the fact that time to failure for the unnotched specimen corresponds to the time when the specimen yielded by necking whereas the time to failure for the notched specimen is when the specimen completely fractures. In essence, the notch plays practically no part in changing the time for ductile failure except that it changes the cross-section area of the specimen. On the other hand the notch is necessary for initiating the crack growth that causes brittle failure.

It is interesting to have an overview of the fractured surface as the stress changes from the ductile to the brittle region. Fig. 11 shows the fractured surface. In general the smooth area associated with slow crack growth increases as the stress decreases. In the fully ductile region no evidence of slow crack growth is evident and the extremely coarse elongated fibres are indicative of the yield process which lead to the ultimate

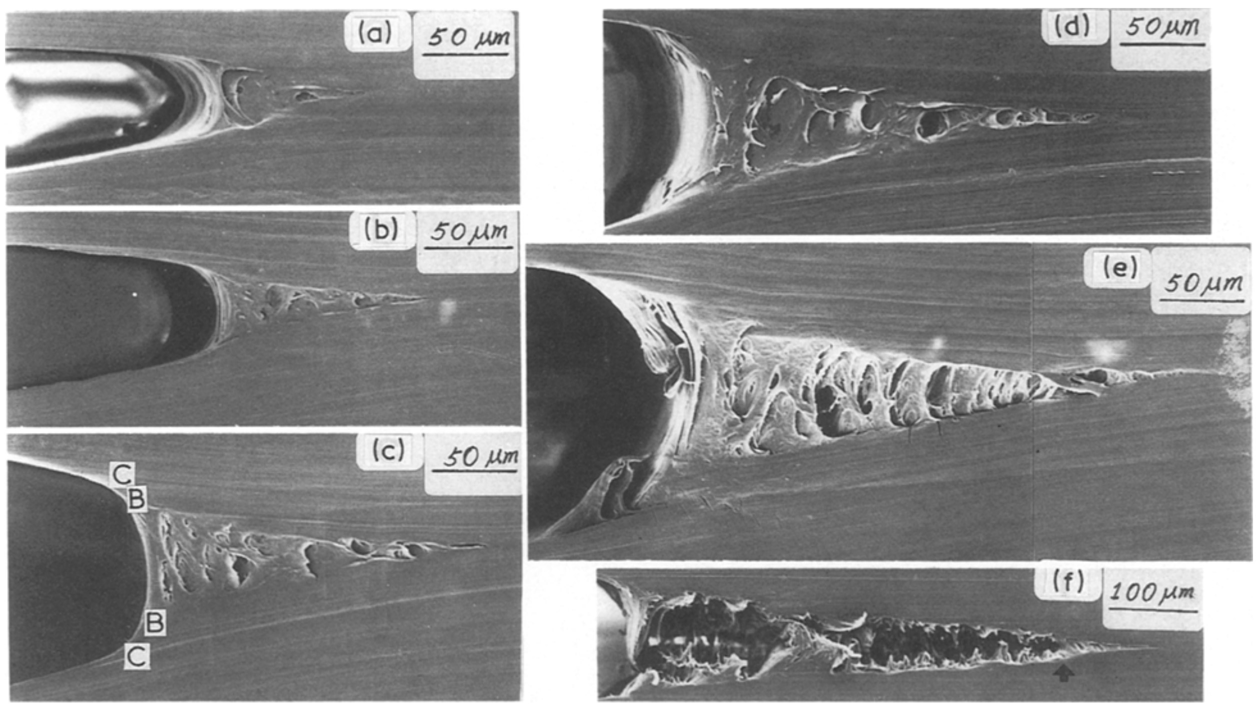

Figure 5 Morphological changes by SEM in notch tip damage zone in brittle region of failure with loading time. The letters a to f correspond to the times indicated in Fig. 4.

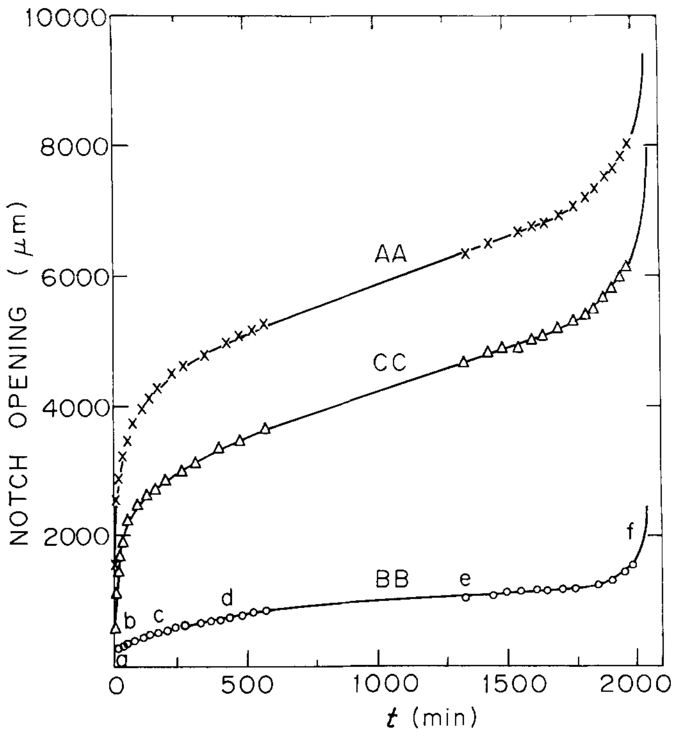

Figure 6 Notch openings AA, CC, and BB plotted against time in ductile region of failure, tested at 4 MPa and 80° C.

fracture. In the regions of slow crack growth the fibrous structure becomes finer and finer as the stress decreases because the thickness of the craze decreases with stress. The craze is the precursor of slow crack growth.

4. Discussion

The microstructural changes at the root of the notch indicate whether the ultimate fracture is likely to be brittle or ductile. Specifically, the difference between the thickness of the craze and notch opening at the root of the notch is small in the brittle region, large in the ductile region and intermediate in the transition region when compared to the notch opening at the surface of the specimen. By measuring these three notch openings, it is easy to determine, at the very beginning of the test, whether the ultimate failure will be ductile, brittle, or transitional.

In general, for all polyethylenes, the most definitive and most widely used criterion for determining whether the failure's ductile, brittle, or transitional is based on the stress against time to failure curves (Fig. 3) where the time to failure varies as

$$t_f \sim \sigma^{-n}$$

If n is appreciably greater than about 20, the behaviour is ductile, if n is definitely less than about 6 the behaviour is brittle. If n is negative so that t_f increases as σ increases, the failure mode is in the transition region. Often investigators report their results as being in the brittle region because the failure is of the slit mode. However, failure in the transition region is also

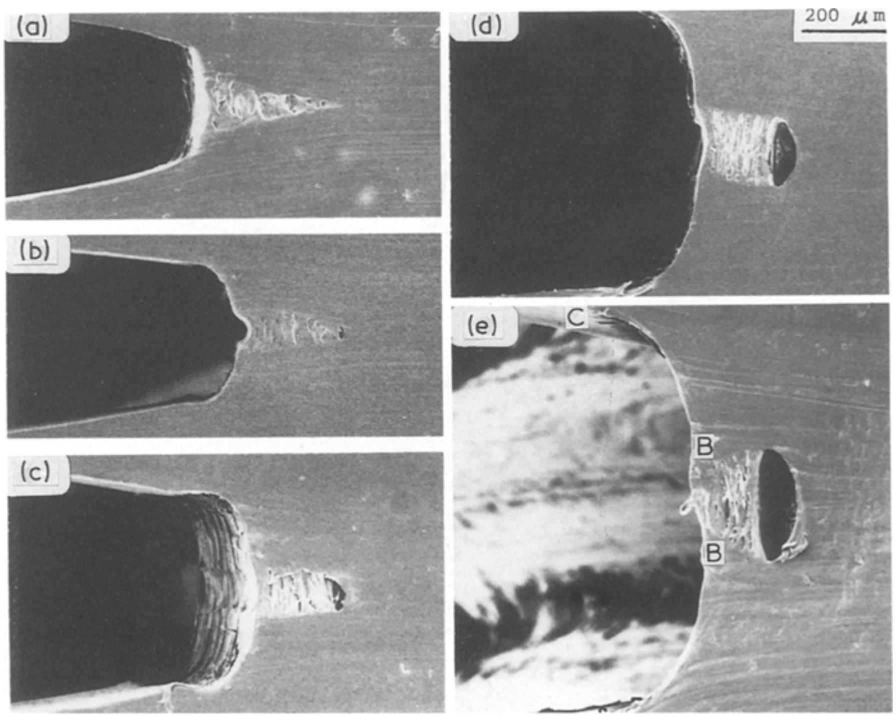

Figure 7 Changes in morphology by SEM of notch tip damage zone in ductile region of failure with loading time. The letters a to e correspond to the times indicated in Fig. 6.

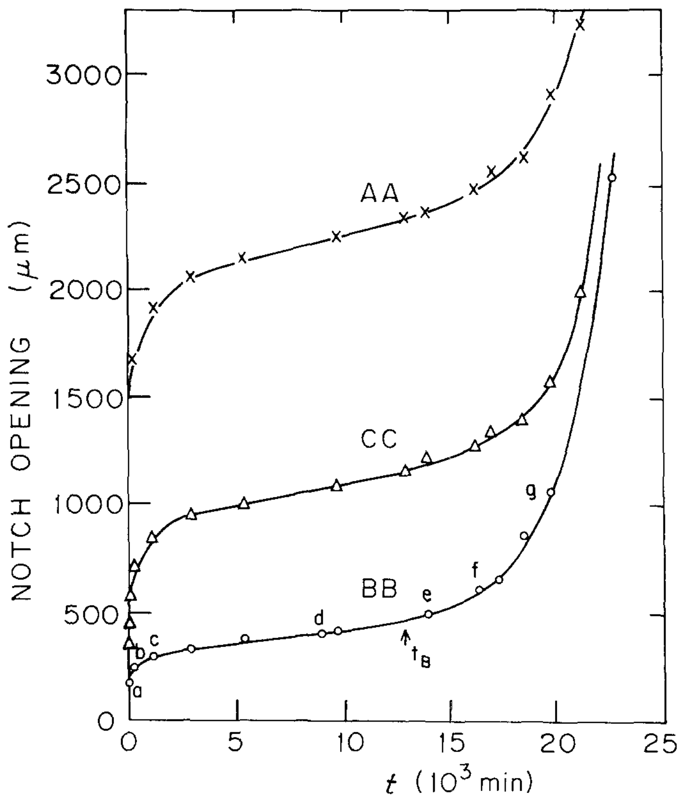

Figure 8 Notch openings AA, CC, and BB plotted against time in the transitional region of failure, tested at 3.2 MPa and 80° C.

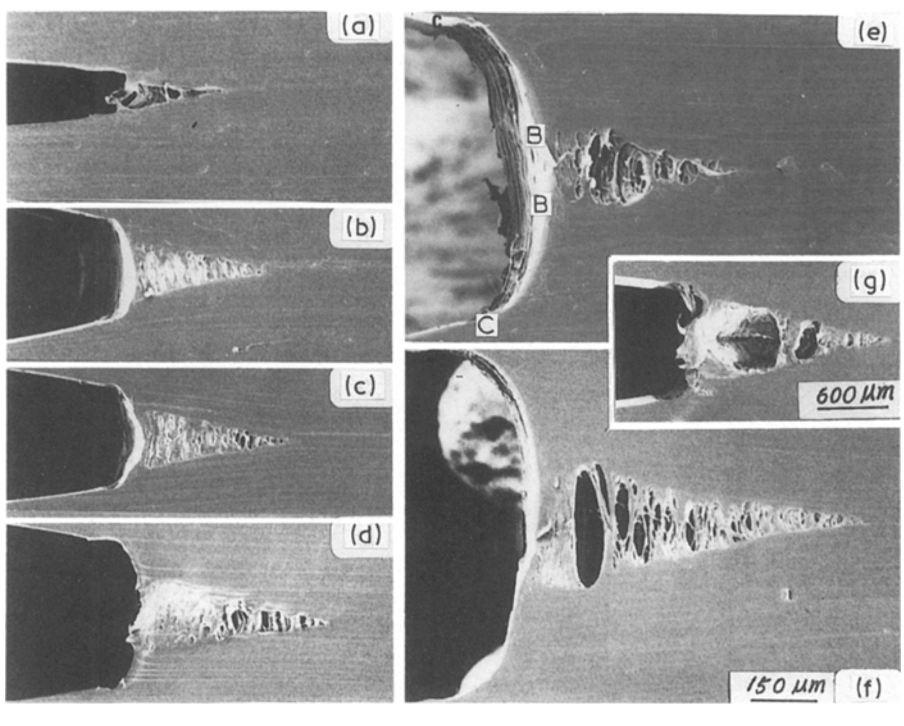

Figure 9 Changes in morphology by SEM of notch tip damage zone of copolymer polyethylene in the transitional region of failure with loading time. The letters a to g correspond to the times in Fig. 8.

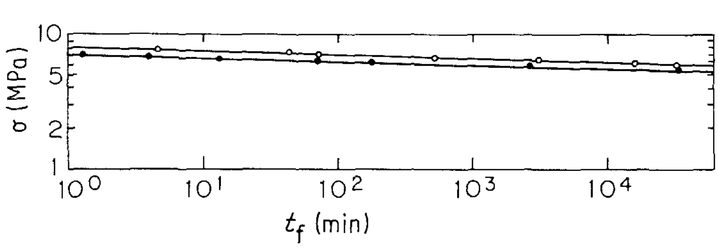

Figure 10 Stress plotted against time to failure in the ductile region for notched (○) and unnotched (●) specimens. The notched specimen fracture failure, ligament stress; unnotched specimen yield failure.

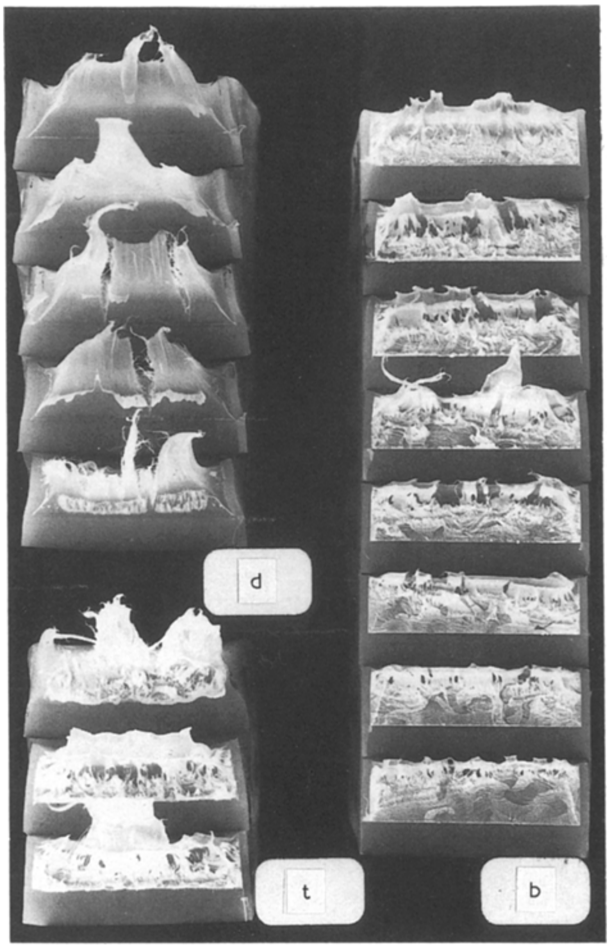

Figure 11 General features of the fracture surface as a function of stress. In the ductile region, d, 4.6 to 3.8 MPa. In the transitional region, t, 3.4 to 3.0 MPa. In the brittle region, b, 2.8 to 1.4 MPa.

of the slit mode. Since the transition region is fairly narrow, this lack of recognition of the difference between the transition and brittle region is usually not important from a practical viewpoint. However, if one wishes a complete and fundamental view of the slow crack growth behaviour of various types of polyethylene resins, then it is important to distinguish between the ductile, brittle, and transitional mode of failure.

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