

# The correlation of slow crack growth in linear polyethylene by the $J$ -integral

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The fracture of a polyethylene (PE) homopolymer by slow crack growth was measured with a three point bending (TPB) specimen and a single edge notch tension (SENT) specimen. The crack growth rate correlated with the stress intensity for each type of specimen. However, for a given  $K$ , SENT specimens exhibited a faster crack growth rate than TPB specimens. Since PE is non-linear, the  $J$ -integral is more appropriate than  $K$  and it was found that  $J$  does correlate the SENT and TPB results. In addition, the degree of non-linearity of the PE was increased by quenching. For the quenched state it was also found that  $J$  correlated the SENT and TPB results in accordance with the dependence of  $J$  on the degree of non-linearity of the material.

(Keywords: polyethylene; slow crack growth rate; non-linear deformation;  $J$ -integral)

## INTRODUCTION

Slow crack growth in polyethylene (PE) occurs by small scale yielding as shown by the microstructure observations by Lu and Brown<sup>1-3</sup>. In experiments with single edge notch tension specimens (SENT) it was found that the stress intensity factor,  $K$ , correlated the effects of stress and notch depth. Similar results were also found for three point bending (TPB) specimens. However, the rate of damage for the SENT and TPB specimens did not coincide for the same value of the stress intensity factor. Since PE exhibits a non-linear elastic stress-strain behaviour before yielding, Bassani *et al.*<sup>4</sup> calculated the  $J$ -integral for these specimens and showed that the  $J$ -integral correlated the SENT and TPB data for slow cooled material.

The  $J$ -integral can correlate the initial rate of damage and, possibly, crack growth for different specimen geometries and loading, but its value depends on the degree of non-linear behaviour in the material. In this investigation the degree of non-linearity of the PE was increased relative to the previously slow cooled material by quenching from the melt. It is the purpose of this paper to determine whether  $J$  also correlates behaviour between the SENT and TPB specimen in PE for the quenched state and to determine how the difference between the  $J$ -integrals for a given rate of damage varies between the slow cooled and quenched materials.

There is a simple physical explanation to show why, for a given rate of damage,  $K$ (SENT) is not equal to  $K$ (TPB), as discussed by Bassani *et al.*<sup>4</sup>. See Figure 1 for the meaning of symbols. The stress intensity factor can be expressed as:

$$K = F(a/b)\sigma_N(\pi a)^{1/2} \quad (1)$$

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where  $\sigma_N$  denotes the nominal stress, and  $F(a/b)$  is, from Reference 5, given by

$$F(\text{SENT}) = 1.12 - 0.231(a/b) + 10.55(a/b)^2 - 21.72(a/b)^3 + 30.39(a/b)^4 \quad (2)$$

and for  $L/b = 4$

$$F(\text{TPB}) = 1.107 - 2.120(a/b) + 7.71(a/b)^2 - 13.55(a/b)^3 + 14.25(a/b)^4 \quad (3)$$

In the range  $0.03 \leq a/b \leq 0.1$ ,  $F(\text{SENT}) = 1.15 \pm 0.03$  and  $F(\text{TPB}) = 1.00 \pm 0.03$ . That is, for the shallow edge cracks in this investigation  $K$  is approximately proportional to  $\sigma_N a^{1/2}$  for each specimen, although the proportionality factor differs.

Now, the stress for the SENT specimen is defined as

$$\sigma_N(\text{SENT}) = P_T/bW \quad (4)$$

This estimate is valid for both linear and non-linear

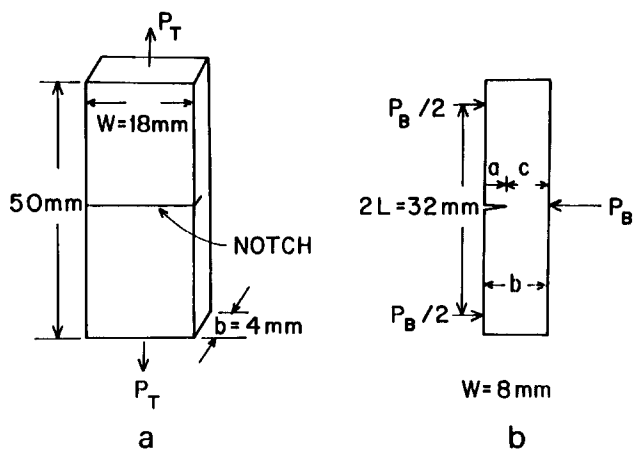


Figure 1 (a) SENT geometry; (b) TPB geometry

material behaviour. For a linear elastic material the outer fibre stress in the beam is

$$\sigma_N(\text{TPB}) = 3PL/Wb^2 \quad (5)$$

On the other hand, for a non-linear material whose stress-strain behaviour is described by a power law:

$$\varepsilon = \varepsilon_0(\sigma/\sigma_0)^n \quad (6)$$

the outer fibre stress is easily calculated to be

$$\sigma_N(\text{TPB}) = (n^{-1} + 2)PL/Wb^2 \quad (7)$$

Here  $n$  is the parameter that determines the degree of non-linear behaviour in the material and  $\varepsilon_0$  and  $\sigma_0$  are material constants.

Since  $n > 1$  for a non-linear material, the outer fibre stress in the non-linear material is less than that in the linear material for the same beam geometry and load. For shallow edge cracks, this outer fibre stress governs the intensity of the crack tip stress. Thus, the  $K(\text{TPB})$  which is based on the linear calculation is greater than the actual stress intensity for the non-linear material. Of course, in dealing with the non-linear material,  $K$  is not applicable and theory suggests the  $J$ -integral<sup>4,5</sup> as a correlating parameter. The  $J$ -integral did correlate the SENT and TPB data for slow cooled material and it is the purpose of this investigation to determine whether the  $J$ -integral would still correlate the data if  $n$  was increased by using quenched PE. We emphasize that the non-linear elastic behaviour referred to is in the bulk of the specimen and typically corresponds to a small  $n > 1$ . Deformation at the crack tip is likely to be highly non-linear.

### FRACTURE PHENOMENON AND CORRELATION

We will now describe the fracture phenomenon that is being investigated. PE under low stresses and around room temperature fails in a macroscopically brittle manner by a process of slow crack growth. In extensive investigations of the kinetics of this fracture process, Lu and Brown<sup>1-3</sup> measured the crack opening displacement at the root of a notch as a function of time under a constant load. They found that the entire curve, including the time to failure, was governed by the initial slope of the curve, which we call  $\delta_0$ . The lifetime  $t_f$  is approximately given, therefore, by

$$t_f = (a_0 + \delta_c)/\delta_0 \quad (8)$$

where  $a_0$  is the initial notch depth and  $\delta_c$  is a constant. Thus,  $\delta_0$  is the essential fracture parameter, whose correlation by fracture mechanics is of primary interest.

In the previous investigations on slow cooled polyethylene by Lu and Brown<sup>1-3</sup>,  $\delta_0$  was related to the stress and initial notch depth at 42°C as follows:

$$\delta_0(\text{SENT}) = 3.1 \times 10^{-10} \sigma_N^{5.0} a_0^{1.9} (\mu\text{m min}^{-1}) \quad (9)$$

$$\delta_0(\text{TPB}) = 3.1 \times 10^{-11} \sigma_N^{5.3} a_0^{1.8} (\mu\text{m min}^{-1}) \quad (10)$$

where  $\sigma_N$  is in MPa and  $a_0$  in  $\mu\text{m}$ .

When  $\log \delta_0$  was plotted against the initial stress intensity,  $K_0$ , it was found that the best fit is:

$$\delta_0(\text{SENT}) = 76K_0^{4.7} (\mu\text{m min}^{-1}) \quad (K \text{ in MPa m}^{1/2}) \quad (11)$$

$$\delta_0(\text{TPB}) = 35K_0^{4.6} (\mu\text{m min}^{-1}) \quad (12)$$

where the correlation coefficient is 0.96. The detailed dependence of  $\delta_0$  on  $\sigma_N$  and  $a_0$  in equations (9) and (10)

does not exactly conform to a simple dependence on stress intensity,  $K_0 \propto \sigma_N(a_0)^{1/2}$ , i.e. the exponent of  $a_0$  is not exactly half the exponent of the stress. Nevertheless, the correlation of  $\delta_0$  with  $K_0$  is still very good, as shown in Figure 8.

In this paper it will be shown that the  $J$ -integral is the most appropriate correlation parameter between SENT and TPB specimens for quenched PE, as was the case for the slow cooled material. Owing to different degrees of non-linearity, the correlations for each material are distinct.

### EXPERIMENTS

The PE used was Marlex 6006 made by Phillips Chemical Company. It is a linear PE with  $M_n = 19\,800$  and  $M_w = 130\,000$ . The density of the slow cooled (SC) state was 0.964 and of the quenched (Q) state 0.941. The yield points at 300 K and at a strain rate of  $0.3 \text{ min}^{-1}$  were 32 and 27 MPa for the SC and Q state, respectively. Their stress curves are shown in Figure 2. The resin was compression moulded into  $4.3 \times 160 \times 160 \text{ mm}^3$  plaques in a close fitting die and these were exposed to about three cycles of pressure between 0 and 6.6 MPa during melting in order to obtain the most consistent material. The SC state was cooled in the press overnight and the Q state was obtained by quenching the die in water. The Q state was annealed at 80°C for 4 days to relax the residual stresses.

The shallow-crack SENT and TPB specimen geometries with mode I loadings are shown in Figure 1. Notch depths varied from 0.18 to 0.43 mm so that the ratio of initial notch length to specimen width,  $a_0/b$ , varied from 0.03 to 0.1. The specimen thickness ( $W$ ) was 18 mm for the SENT specimen and 8 mm for the TPB specimen. Previous work showed that the resulting fracture was almost completely plane strain for these geometries. The stresses were chosen so that they were less than one-half the yield point in order to produce a planar deformation zone as shown by Bhattacharya and Brown<sup>6</sup>. For the SC state the stresses ranged from 7.5 to 12 MPa and for the Q state from 7 to 10.5 MPa. Altogether, about 75 specimens were tested, all at  $42 \pm 0.2^\circ\text{C}$ .

The crack tip opening displacement (COD),  $\delta$ , was measured with a filar eyepiece in an optical microscope at a magnification of 100 by looking directly into the crack. The error in measuring  $\delta$  was about  $\pm 2 \mu\text{m}$ .

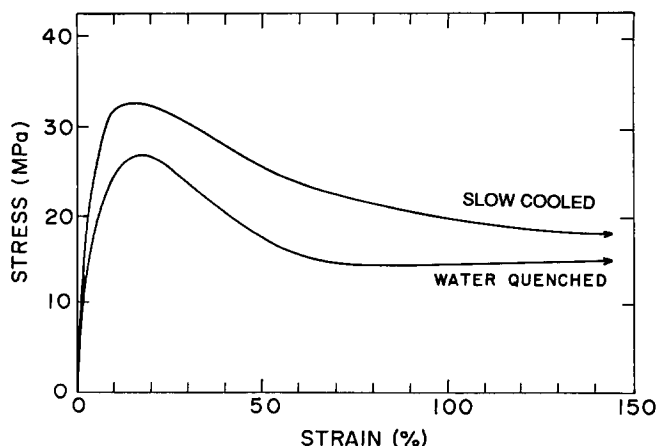


Figure 2 Stress-strain curves of SC and Q states

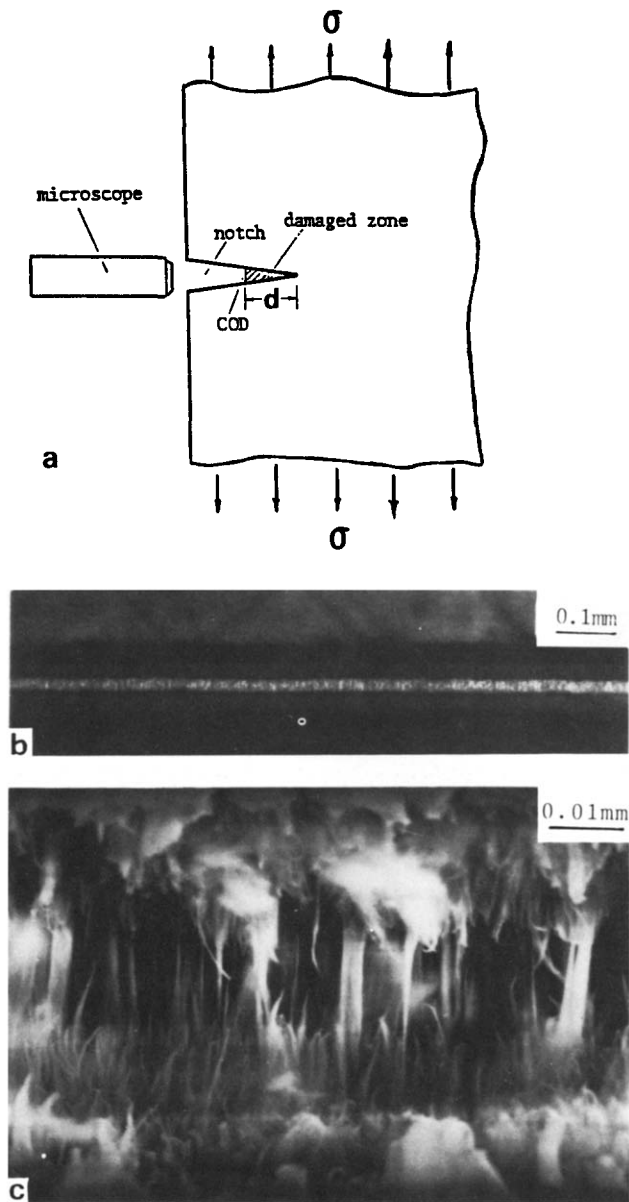


Figure 3 Experimental set-up for measuring COD

Figure 3 shows a view of the COD as seen through the microscope. The opening of the notch at the specimen surface and the macroscopic creep strain were also measured.

Details of the damaged zone have also been observed by taking slices from the interior of a test specimen and viewing them in the SEM while the crack was held open in a jig the same amount as occurred during the test (Figure 4). The ratio of  $\delta$  to the length of the damage zone was equal to about 1/7 during most of the test except for the late stage of crack growth when it increased.

EXPERIMENTAL RESULTS

Figure 5 shows a typical test result. In general, the following behaviour was observed:

- (1) the initial loading produced an instantaneous COD,  $\delta_0$ ;
- (2) the slope of the  $\delta$  versus  $t$  curve,  $\dot{\delta}$ , remained constant up to the beginning of crack growth, as indicated by the solid tangent line;
- (3) after crack growth started  $\dot{\delta}$  increased continuously

until rapid fracture occurred. The macroscopic creep strain was generally <8%.

Note that the difference between the openings of the crack at the surface and at the crack tip is constant during much of the first part of the test. This is consistent with the microscope observations (Figure 4) that the ratio of  $\delta$  over the length of the damage zone is also constant during the same time interval. Figure 6 shows a series of  $\delta$  versus  $t$  curves for a range of stresses. These curves are typical for both materials and loading modes and for variations in the depth of the initial notch.

The differences between the two materials and the two loading methods are shown in Figure 7 where  $\delta$  versus  $t$  curves are shown for the same value of the stress intensity. The important aspect of these curves is the initial slope,  $\dot{\delta}_0$ , which essentially governs the failure process. As shown previously by Lu and Brown<sup>3</sup>, the Q state fractures faster than the SC state and the SENT specimens fracture faster than the TPB specimens. In addition, the ratio  $\dot{\delta}_0(\text{SENT})/\dot{\delta}_0(\text{TPB})$  for the Q state is greater than that for the SC state. This result is expected since the Q state is more non-linear than the SC state.

Figure 8 shows  $\dot{\delta}_0$  versus  $K_0$  for the four combinations of materials and methods of loading. The relationships for the SC state are given in equations (11) and (12); the relationships for the Q state are:

$$\dot{\delta}_0(\text{SENT}) = 186K_0^{4.7} (\mu\text{m min}^{-1}) \quad (13)$$

$$\dot{\delta}_0(\text{TPB}) = 67K_0^{4.7} (\mu\text{m min}^{-1}) \quad (14)$$

The correlation coefficients are all about 0.96. For all conditions it is seen that the exponential dependence on  $K_0$  is the same, namely 4.7. The pre-exponential factor contains the information concerning the behaviour of the SC state vis-à-vis the Q state and SENT specimens vis-à-vis TPB specimens. It is seen that  $\dot{\delta}_0(\text{SENT})/\dot{\delta}_0(\text{TPB})$  is 2.8 for the Q state and 2.2 for SC state. This is consistent with the greater non-linear behaviour of the Q state.

J-INTEGRAL ANALYSIS

Since PE is a non-linear elastic material the J-integral is the appropriate parameter rather than K (ref. 7). J will now be determined for each state and loading geometry. Solutions for J for various geometries and values of n are given in Reference 8. For plane strain conditions:

$$J(\text{TPB}) = \sigma_0 \epsilon_0 c h(a/b, n) (P/P_0)^{n+1} \quad (16)$$

$$P_0 = 0.728 \sigma_0 c^2 / L$$

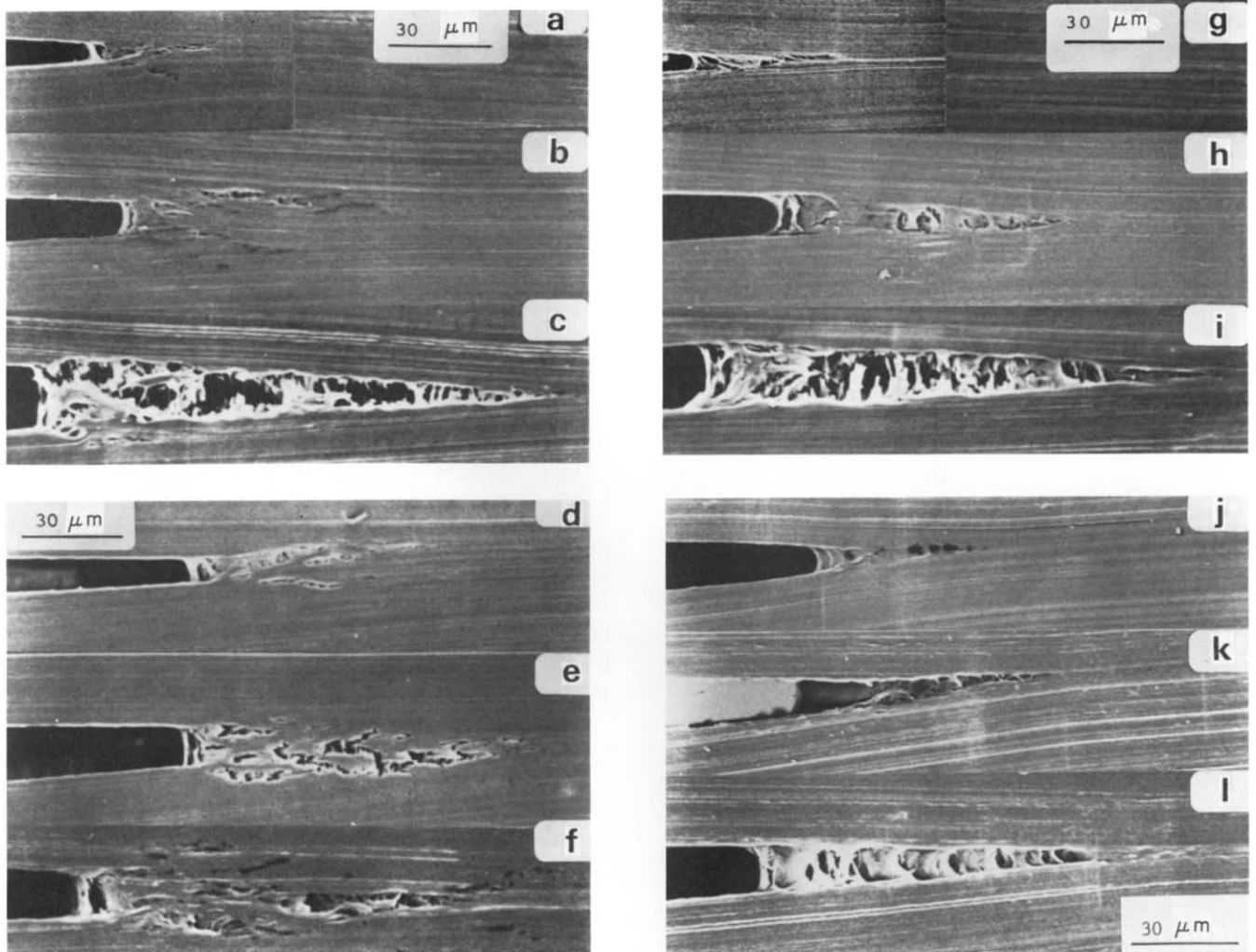
where L, a, b and c are defined in Figure 1,

$$J(\text{SENT}) = \sigma_0 \epsilon_0 c (a/b) h(a/b, n) (P/P_0)^{n+1} \quad (17)$$

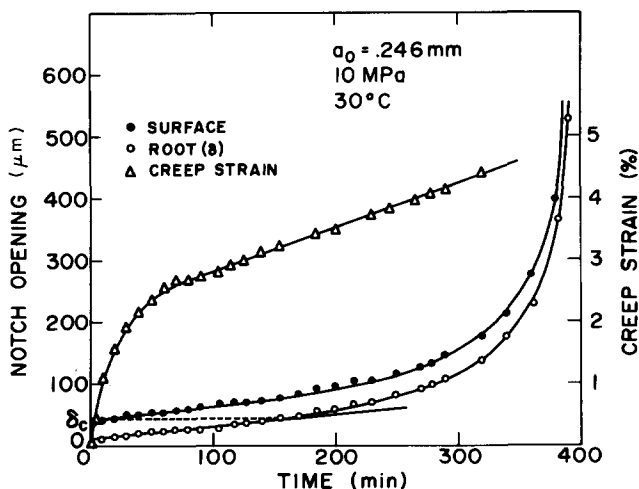
where  $P_0 = 1.455 \{ [1 + (a/c)^2]^{1/2} - a/c \} \sigma_0 c$ , and  $\sigma_0$ ,  $\epsilon_0$  and n are from equation (6).  $h(a/b, n)$  is a function of the a/b ratio and n. P is the load per unit thickness. Values of  $h(a/b, n)$  are tabulated in Reference 8 for the SENT and TPB specimens over a range of a/b and n values. In these experiments a/b ranged from 0.03 to 0.1. The smallest value of a/b that is given in Reference 8 is 0.125, so that the data required extrapolation to the smaller a/b values. However, He and Hutchinson<sup>9</sup> give a complete solution for a/b → 0. Note also that for a linear solid, n=1, under plane strain conditions

$$J = (1 - \nu^2) K^2 / E \quad (18)$$

where E is Young's modulus and  $\nu$  is Poisson's ratio.

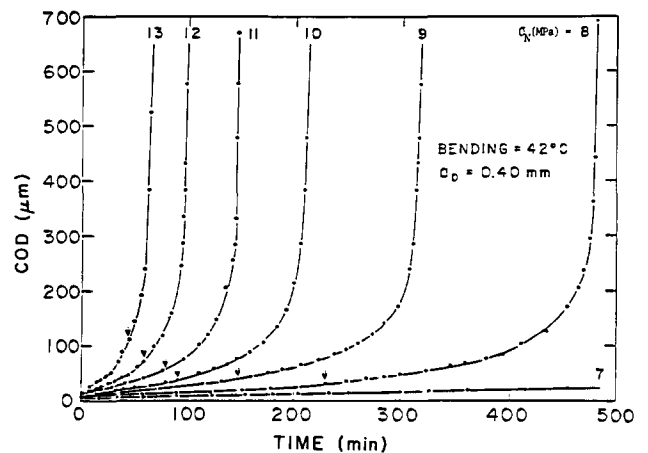


**Figure 4** SEM micrographs of (a)–(f) slow cooled specimens and (g)–(l) quenched specimens. (a)–(c) TPB after (a)  $t=6.4$  min,  $K=0.4 \text{ MPa m}^{1/2}$ ; (b) 25 min,  $0.40 \text{ MPa m}^{1/2}$ ; (c) 65 min,  $0.40 \text{ MPa m}^{1/2}$ . (d)–(f) SENT after (d) 5.6 min,  $0.5 \text{ MPa m}^{1/2}$ ; (e) 22 min,  $0.36 \text{ MPa m}^{1/2}$ ; (f) 22 min,  $0.42 \text{ MPa m}^{1/2}$ . (g)–(i) TPB after (g) 2 min,  $0.40 \text{ MPa m}^{1/2}$ ; (h) 3.8 min,  $0.40 \text{ MPa m}^{1/2}$ ; (i) 18 min,  $0.3 \text{ MPa m}^{1/2}$ . (j)–(l) SENT after (j) 1.3 min,  $0.40 \text{ MPa m}^{1/2}$ ; (k) 3 min,  $0.40 \text{ MPa m}^{1/2}$ ; (l) 6.5 min,  $0.41 \text{ MPa m}^{1/2}$



**Figure 5** Typical COD versus time and creep versus time curves

Equations (16) and (17) were evaluated for the various values of  $a$ ,  $b$  and  $P$  that were used in the experiments. The choice of  $n$  depends on the stress–strain curves (Figure 2). Log–log plots of the stress–strain curves (Figure 9) show that for PE the actual stress–strain curves (Figure 2) only approximate the power law relationship



**Figure 6** COD versus time for various stresses, for TPB for slow cooled specimen

(equation (6)) and that the values of  $n$  range between 1.1 and 3.5. Therefore  $J$  was evaluated for several values of  $n$ , and the value of  $n$  which best collapsed the SENT and TPB data is taken as the most appropriate value. In Figure 10,  $\delta_0$  versus  $J$  was calculated for the Q state with  $n=1.5$  and 2.0. The value of  $n=2$  nicely collapses the data within the experimental scatter. Figure 11 shows

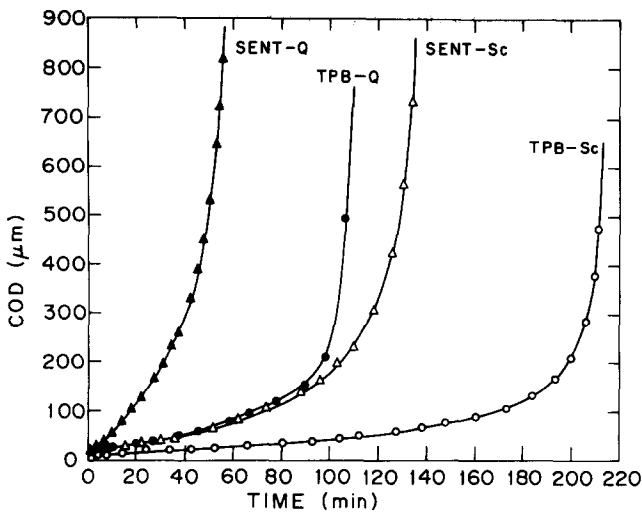


Figure 7 COD versus time for Q(SENT), Q(TPB), SC(SENT) and SC(TPB) for  $K=0.4 \text{ MPa m}^{1/2}$  and notch depth = 0.4 mm

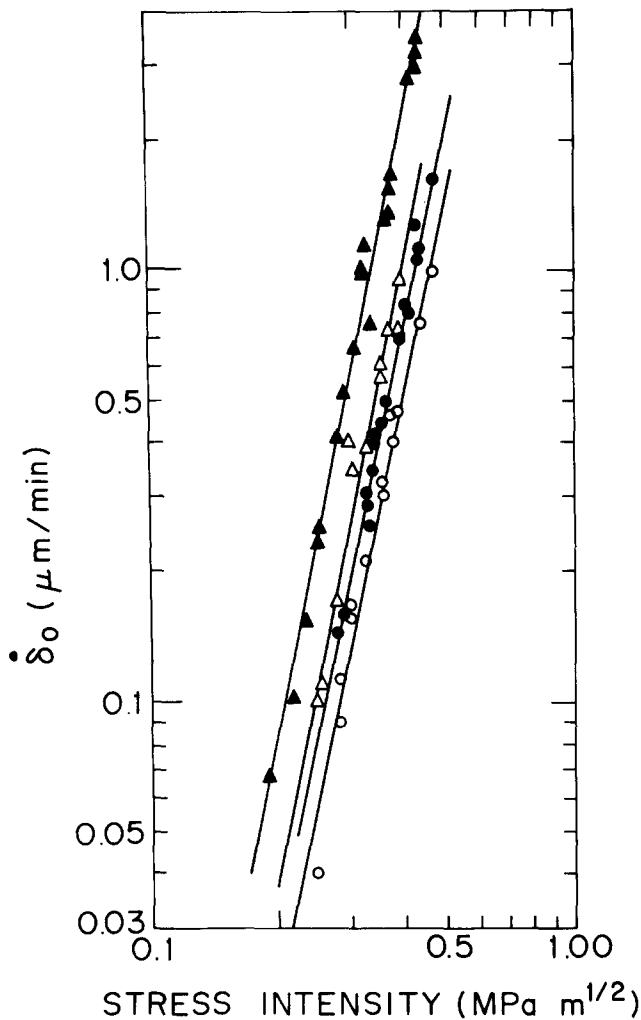


Figure 8  $\delta_0$  versus  $K$  for:  $\blacktriangle$ , Q(SENT);  $\triangle$ , SC(SENT);  $\bullet$ , Q(TPB); and  $\circ$ , SC(TPB)

that for the SC state the best value is  $n=1.5$ . That  $n$  for the SC state is less than  $n$  for the Q state is consistent with Figure 9, which shows that the SC state is less non-linear than the Q state.

In Figures 10 and 11  $\delta_0$  is plotted versus  $J'$ , a normalized value of  $J$ :

$$J' = J/\sigma_0 \epsilon_0 b \quad (18)$$

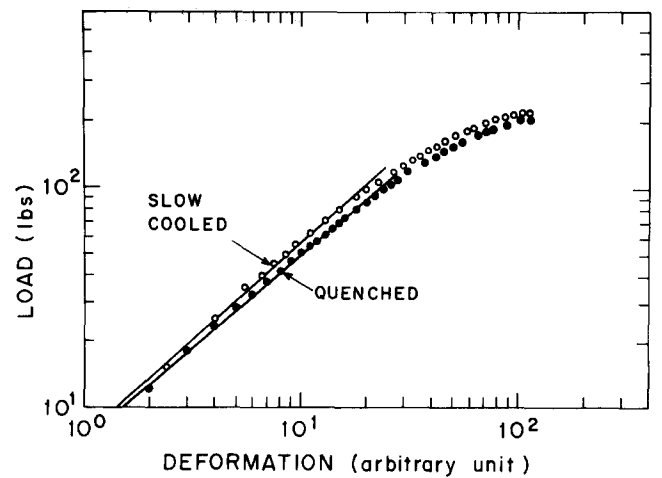


Figure 9 Log stress versus log strain obtained by using Figure 2

where  $\sigma_0$  and  $\epsilon_0$  were taken to be the yield stress and yield strain of the material and  $b$  is a characteristic dimension of the specimen, which in this case is the width of the specimen. This normalization is simply a convention to obtain a non-dimensional measure for the  $J$ -integral. From Figures 10 and 11 the following relationships are found:

$$\delta_0(\text{SC}) = 2 \times 10^3 J'^{1.8} \text{ (}\mu\text{m min}^{-1}\text{)} \quad (19)$$

$$\delta_0(\text{Q}) = 4 \times 10^3 J'^{1.8} \text{ (}\mu\text{m min}^{-1}\text{)} \quad (20)$$

It is interesting to note that the dependence of  $\delta_0$  on  $J'$  is the same for both states.  $J'$  is related to the strain energy release rate, which is related to the energy per unit area of the damage zone. The coefficient  $2 \times 10^3$  for the SC state and  $4 \times 10^3$  for the Q state comes from the time dependence of the strain energy release rate on the morphology of the polymer and on the temperature. Previous work shows that  $\delta_0$  varies as  $e^{-Q/RT}$ , where  $Q$  is about the same for the Q and SC states. Therefore the difference between the coefficients stems from the difference in morphology between the SC and Q state. The fact that  $\delta_0$  varies as  $J'^{1.8}$  is not understood at this time.

It is of interest to calculate the absolute values of  $J$  that operated in these experiments and to compare them with  $J_c$  as measured by a room temperature rapid fracture experiment on PE. An absolute value of  $J$  is obtained by evaluating equations (16) or (17). These equations involve three components:

- (1) a geometric one that involves the geometric factors  $a$ ,  $b$ ,  $c$ ,  $L$  and  $W$ ;
- (2) the load  $P$ ; and
- (3) the material parameters  $n$ ,  $\sigma_0$  and  $\epsilon_0$ .

Since the material is assumed to obey a power law as given by equation (6), there are really only two independent material parameters  $n$  and  $(\sigma_0/\epsilon_0^{1/n})$ . These material parameters are obtained by best-fitting equation (6) to the actual stress-strain curves in Figure 2 for the value of  $n$  which best collapses the data, as shown in Figures 10b and 11, where  $n=2$  and  $n=1.5$  are best for the Q and SC states, respectively. Using these values of  $n$ , the best fit of equation (6) to the actual stress-strain curves in Figure 2 gives  $(\sigma_0/\epsilon_0^{1/n})$  values of 67 and 120 MPa for the Q and SC materials, respectively. After inserting the above values into equations (16) and (17) for the various values of  $a$ ,  $b$ ,  $c$  and  $P$  it was found that the values of  $J$  that were operating during these slow crack growth

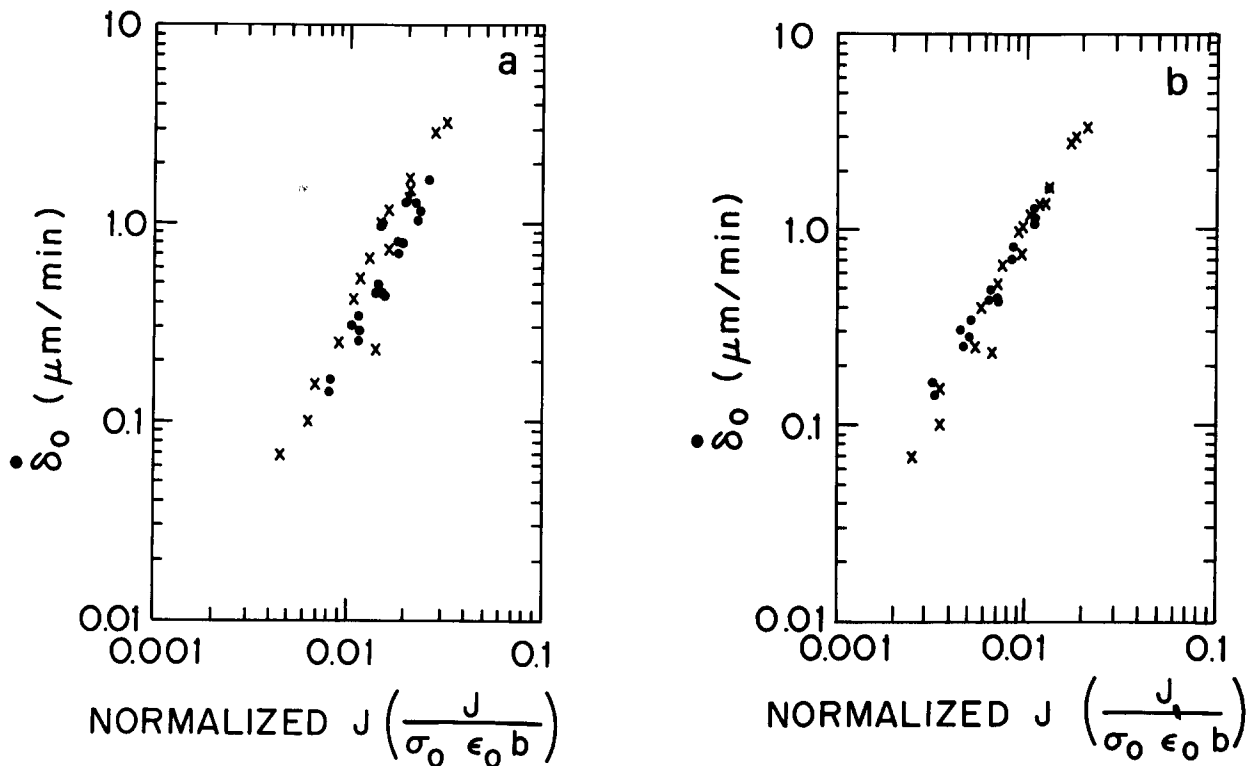


Figure 10  $\delta_0$  versus normalized  $J$  for Q state: (a)  $n=1.5$ ; (b)  $n=2.0$ .  $\times$ , SENT;  $\bullet$ , TPB

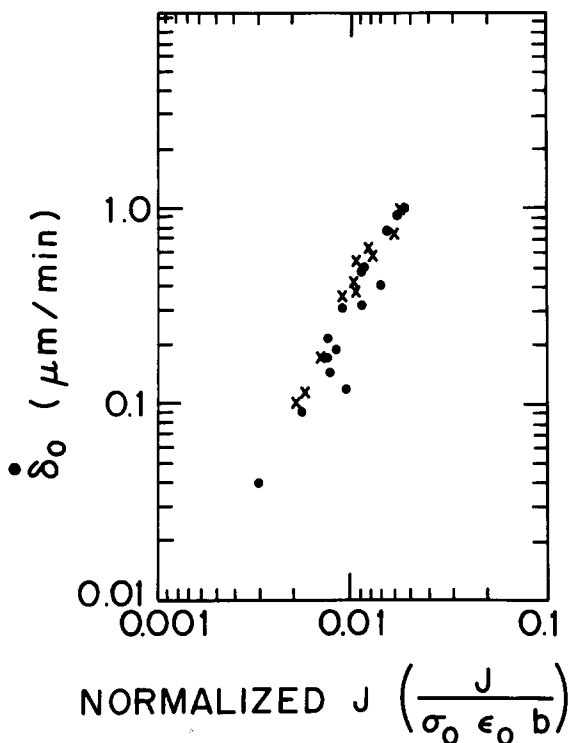


Figure 11  $\delta_0$  versus normalized  $J$  for SC state:  $n=1.5$ ;  $\times$ , SENT;  $\bullet$ , TPB

experiments ranged from about 40 to 220  $\text{J m}^{-2}$ . These values are much smaller than the values of 1000–30000  $\text{J m}^{-2}$  presented by Williams<sup>10</sup> for rapid fracture at 20°C. A recent paper by Qian *et al.*<sup>11</sup> showed that the threshold value of  $J$  for slow crack growth is about 5  $\text{J m}^{-2}$ .

The question arises as to why a kinetic process as represented by the parameter  $\delta_0$  should depend on a

time-independent parameter such as  $J$  and not on a time-dependent fracture parameter such as  $C^*$ . The answer hinges on the fact that  $\delta_0$  represents the initial rate of the damaged process. The amount of creep strain in the matrix that occurs during the time interval for measuring  $\delta_0$  is small, as shown in Figure 5. The fact that  $J$  works as the correlating parameter for PE with different specimen geometries is because (1) there is small scale yielding, (2) the PE is non-linear, and (3) the creep strain in the specimen is small during the time interval over which  $\delta_0$  is measured. That  $J$  correlates  $\delta_0$  for the homopolymer of linear PE rests ultimately on the experimental evidence as presented in Figures 10 and 11. These experiments may suggest that  $J$  may correlate the slow crack growth behaviour in copolymers of PE which exhibit a much greater crack resistance to a slow crack growth. Experiments to test this suggestion are now in progress.

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