The transition between ductile and slow-crack-growth failure in polyethylene

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In the neighbourhood of room temperature, linear polyethylene will exhibit two types of long-time failure under a constant stress: (a) ductile failure, which is produced by a creep process, and (b) brittle fracture, which is produced by slow crack growth. The kinetics of each failure process have been quantitatively represented based on experimental data. The transition time from one failure mechanism to the other has been calculated in terms of the material parameters relevant to each failure mechanism and the defect responsible for crack growth.

(Keywords: crack growth; failure; polyethylene; fracture)

INTRODUCTION

When polyethylene (PE) fails in a ductile manner it exhibits necking in a tensile test, bulging of a pipe under an internal pressure, or collapse of the pipe wall when bent. Ductile failure occurs under a constant load after a certain time, which depends on the stress and temperature. Under a constant stress, polyethylene may also fail by slow crack growth, which has the macroscopic appearance of a brittle fracture, but microscopically the fracture is ductile and exhibits the appearance of a coarse craze. As in all fracture processes, the crack starts from a point of stress concentration in the material.

The time-dependent fracture of PE at room temperature is in keeping with the high-temperature slow-crack-growth fracture under low stress that occurs in other crystalline materials. It should be realized that room temperature for PE is about $0.78T_m$, where T_m is its melting point. In all crystalline solids the molecular or atomic mobility becomes significant above about $0.5T_m$. PE is a semicrystalline polymer containing alternate layers of crystalline and non-crystalline regions. The non-crystalline regions are expected to be rubber-like because it is generally thought that room temperature is above its glass transition temperature T_g (-27°C). Thus, the high-temperature state of the crystalline regions and the rubber-like state of the non-crystalline regions strongly indicate that under low stresses the molecular mobility should be significant in PE and consequently long-time low-stress brittle fracture should be expected.

The ductile–brittle behaviour of PE is of constant interest because PE gas pipes are being tested prior to installation. The pipe tests, which are based on the ASTM specification D2837, involve measurement of time to failure as a function of stress under an internal hydrostatic pressure. The specification is now being modified to include the effect of temperature. There are several problems connected with these tests: (1) the time to failure in the brittle mode may be extremely long at low temperature below about 50°C; (2) the analysis of the test results is based purely on an empirical evaluation of test data without any fundamental input about the process; (3) the brittle failure is initiated at a random defect, yet there is no attempt to incorporate or measure the contribution of defects. A complete formulation of the ductile–brittle transition requires a knowledge of the stress, mode of loading, temperature and the geometry of the point of stress concentration.

In this paper the failure time is derived from the kinetics of the damage process that leads to failure. We show how the ultimate failure can be predicted from the initial kinetics of damage and thereby obtain results which would otherwise not be available due to the limitations on the length of time that a test can be conducted. A complete analysis of the ductile–slow-crack-growth transition is presented for a linear PE. The approach is sufficiently general so that it is also expected to be useful for predicting the lifetime of other polymers.

EXPERIMENTAL MATERIALS

The commercial resins were obtained from the Phillips Chemical Company with the designations Marlex 6006, which is a linear homopolymer, $M_n = 19\,600$, $M_w = 130\,000$. The pellets were compression moulded and slowly cooled from the melt at a rate of 0.5°C min^{-1}. The specimens were 4.3 mm thick with a density of 0.964 g cm^{-3}. Crack growth was measured in single edge-notched tension and notched three-point bending specimens. The specimens were 8–18 mm wide to ensure that plane strain conditions occurred. Notches were made with a razor under very carefully controlled conditions with depths varying from 0.15 to 0.40 mm along the width of the specimen. The crack opening displacement (COD) was measured with a microscope by viewing the interior of the notch (*Figure 1*). Details are given in previous papers[1,2]. The error in measurements of the COD at the notch tip is within $\pm 2\,\mu$m. The temperature of all tests

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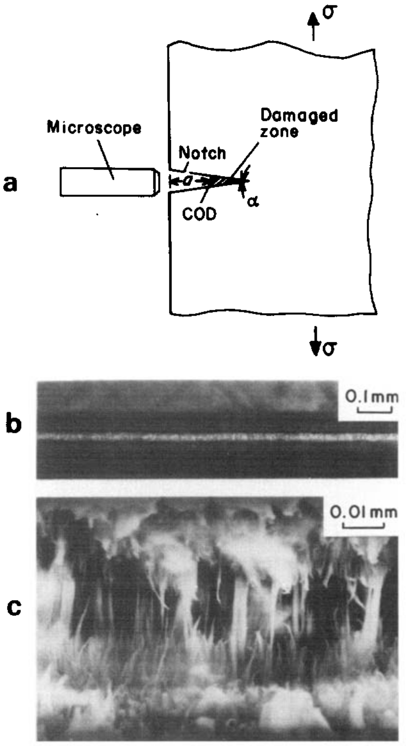

Figure 1 Experimental measurement of COD

was controlled within $\pm 1°C$ and the average temperature was known within $\pm 0.5°C$. The creep specimens had a 25 mm gauge length and the creep strain was measured with a clip-on extensometer.

ANALYSIS OF THE DUCTILE–BRITTLE TRANSITION

Ductile failure

Ductile failure in PE occurs by yielding, which results in a large plastic strain by necking, bulging or kinking depending on the specimen geometry and loading. Ductile failure occurs under a constant stress when the creep strain reaches a critical value, ε_c. Crissman and Zapas[3,4], Hin and Cherry[5] and the present authors have shown that the critical strain for yielding is $\sim 10\%$. It is also known[6] that, in general, all linear polymers yield at strains of about 6–12%.

The experimental form of the creep curve is shown in *Figure 2* and its equation is well represented by

$$\varepsilon = A\sigma^m t^n \qquad (1)$$

where A is an exponential function of the temperature, and m and n are material parameters, which vary slowly with stress. Crissman and Zapas[3,4] have given a more complete representation of the creep equation but equation (1) adequately describes the data and also leads to a simple quantitative description of the ductile–brittle transition. The time for ductile failure, t_D, is derived from (1):

$$t_D = (\varepsilon_c / A\sigma^m)^{1/n} \qquad (2)$$

The temperature dependence of t_D has been measured by Crissman and Zapas[3] and by Cooney[7]:

$$t_D = \frac{D\varepsilon_c^{1/n}\exp(Q_D/RT)}{\sigma^{m/n}} \qquad (3)$$

where D and Q_D are material parameters.

Brittle failure

Long-time brittle fracture occurs under a constant stress and originates at a defect in the material. In order to control the process and describe the phenomenon quantitatively, it is necessary to initiate the failure process at a well defined defect. In our case, the defect is a notch of a definite depth and sharpness. Under a constant stress the following sequence of events occurs: (1) a craze-like plastic zone immediately emanates from the notch; (2) the plastic zone grows; (3) after a certain amount of growth, a crack starts; (4) the crack grows slowly with a plastic zone in front of it until rapid fracture occurs as predicted by Griffith. The growth of the damaged zone *versus* time is shown in *Figure 3* for single edge-notched tension. The notched three-point bending specimens give similar curves for the same value of the J integral[8].

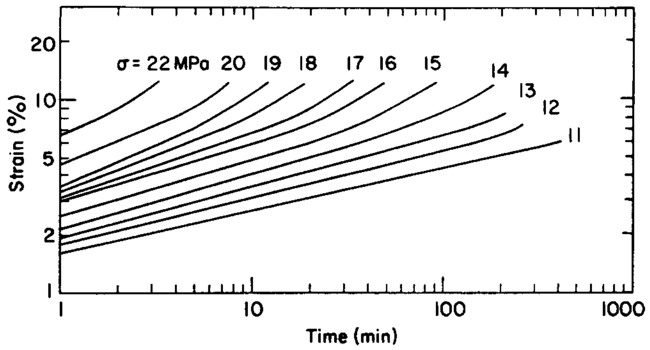

Figure 2 Creep curves at 30°C

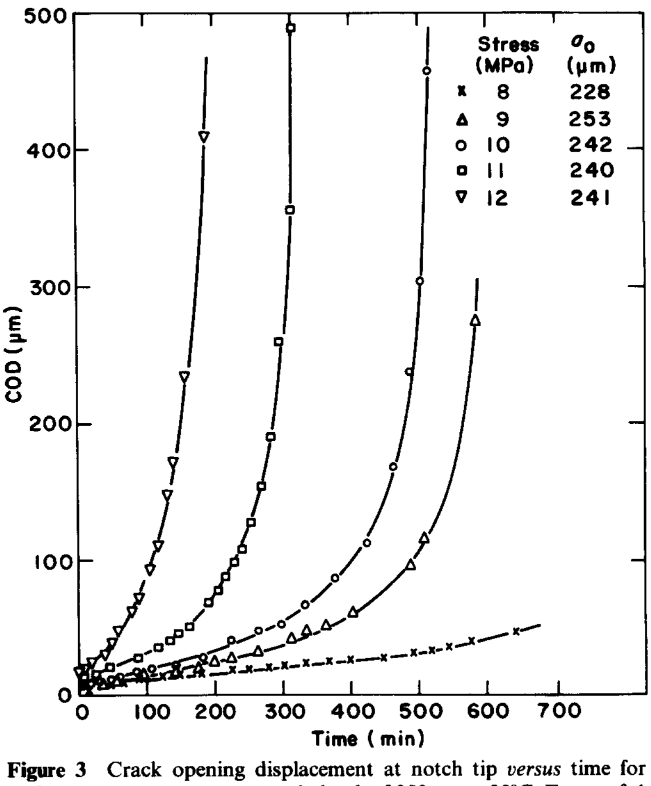

Figure 3 Crack opening displacement at notch tip *versus* time for various stresses at constant notch depth of 250 μm at 30°C. From ref. 1

The microstructure of the damaged zone before and after slow crack growth is shown in *Figure 4*. Note that the damaged zone is triangular in shape. The COD, δ, is the dimension of the damaged zone as measured at the tip of the original notch. Extensive experiments by Lu and Brown[1,2] have shown that the angle of the damaged zone prior to crack growth remains nearly constant and has the value of about 10° for tension and 8° for three-point bending. Thus, measuring δ gives the length of the damaged zone. The damaged zone contains microcrazes and it is the nucleation and growth of these microcrazes that produce the overall growth of the damaged zone.

In *Figures 3* and *5* it is seen that there is an initial COD that occurs when the load is applied. Its size, δ_0, is well described by the Dugdale equation for plane strain conditions:

$$\delta_0 = \frac{K^2}{\sigma_y E(1-v^2)} \tag{4}$$

where K is stress intensity, σ_y is the yield point, E is Young's modulus and v is Poisson's ratio. The damaged zone initially grows at a constant velocity, which is represented by the initial crack opening displacement rate, $\dot{\delta}_0$. At a critical value of the COD, δ_c, crack growth starts; for a single edge-notch tension test $\delta_c = 25$–$30\,\mu$m and for three-point bending $\delta_c = 20\,\mu$m. After crack growth starts the curve of COD vs. t accelerates to complete fracture.

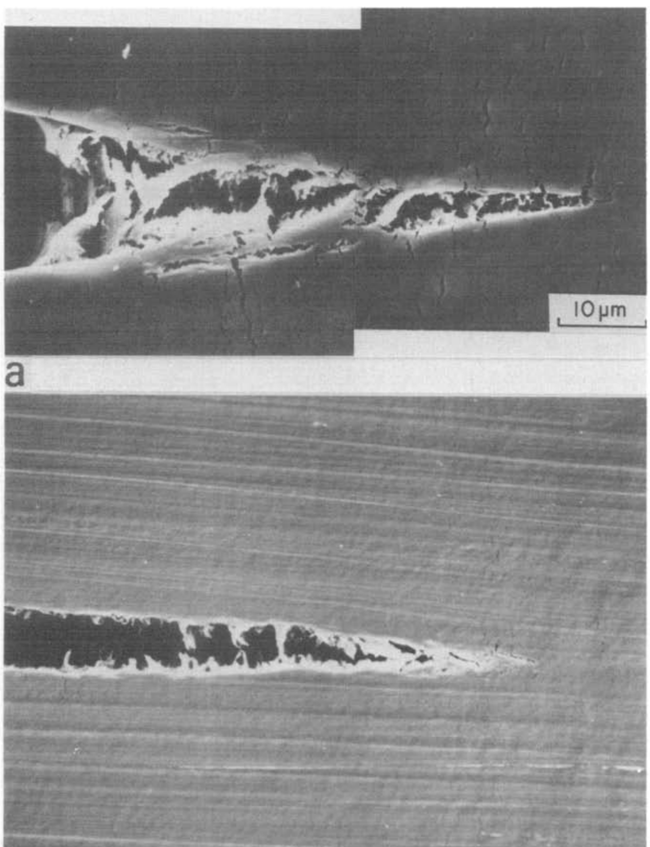

Figure 4 Microstructures of the damaged zone (a) before and (b) after slow crack growth at low stresses. (a) Single edge-notched tension: $a_0 = 257\,\mu$m, $\sigma = 12$ MPa, $T = 30$°C, loading time = 10 min, COD = 20 μm. (b) Single edge-notched tension: $a_0 = 394\,\mu$m, $\sigma = 10$ MPa, $T = 30$°C, loading time = 120 min, COD = 148 μm

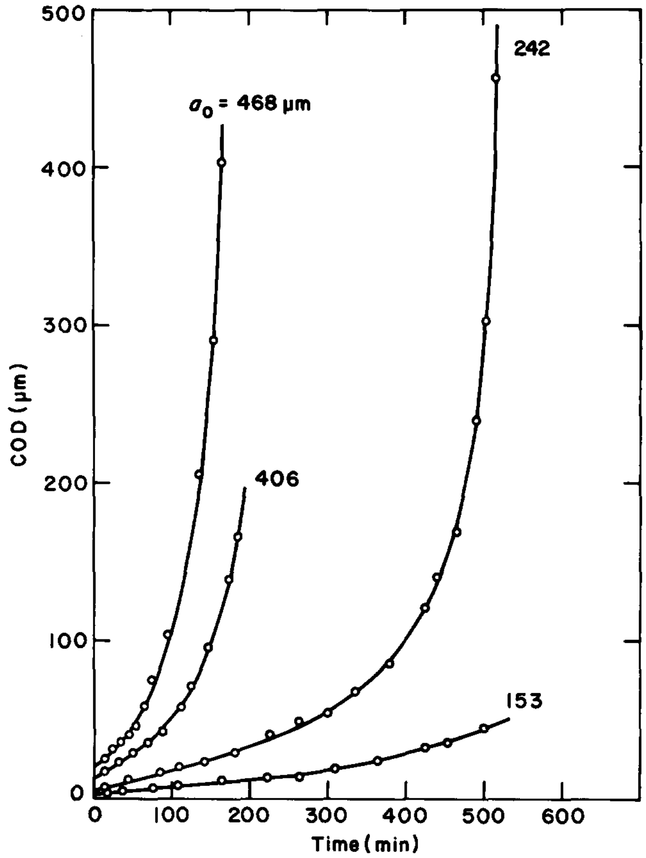

Figure 5 Same as *Figure 3* for various notch depths at a constant stress of 10 MPa. From ref. 1

Lu and Brown[1,2] have shown that the time for complete fracture, t_B, can be predicted from the initial kinetics of the damage process*:

$$t_B = (\alpha a_0 + \delta_c)/\dot{\delta}_0 \tag{5}$$

where α is the ratio of the COD to length of the damaged zone, a_0 is the depth of the initial notch, δ_c is the critical COD for crack growth and $\dot{\delta}_0$ is the initial slope of COD vs. t (*Figures 3* and *5*). Equation (5) is based on the fact that if the initial slope of a curve in *Figures 3* and *5* is known, the slope of the curve, $\dot{\delta}$, beyond δ_c can be calculated from the equation

$$\frac{\dot{\delta}}{\dot{\delta}_0} = \left(\frac{a_0 + \Delta a}{a_0 + \Delta a_c}\right)^2 \tag{6}$$

where $\Delta a = \delta/\alpha$ and $\Delta a_c = \delta_c/\alpha$ as verified by Lu and Brown[1,2].

The dependence of $\dot{\delta}_0$ on T, σ and a_0 has been extensively measured by Lu and Brown[1,2] on Marlex 6006 with the result

$$\dot{\delta}_0 = B\sigma^p a_0^q \exp(Q_B/RT) \tag{7}$$

where B, p, q and Q_B are material parameters, which are about the same for tension and bending except for B. Combining (5) and (7) gives the time for brittle failure:

$$t_B = \frac{(\alpha a_0 + \delta_c)\exp(Q_B/RT)}{B\sigma^p a_0^q} \tag{8}$$

* For three-point bending the prediction is somewhat improved if $\delta_c = 0$

The ductile–brittle transition

Both failure processes begin when the specimen is loaded (*Figure 6*). The ultimate failure mode is determined by whether t_D or t_B is the smaller. The conditions for the ductile–brittle transition are obtained by equation (3) and (8) whereby

$$\frac{\alpha a_0 + \delta_c}{a_0^q} \frac{\exp[(Q_B - Q_D)/RT]\sigma^{(m/n)-p}}{BD\varepsilon_c^{1/n}} = 1 \quad (9)$$

If the left side of (9) is less than 1, the ultimate mode of failure is by brittle fracture.

The time for the ductile–brittle transition $t_{D\text{-}B}$ and the corresponding stress $\sigma_{D\text{-}B}$ are calculated as follows: let

$$t_D = C_D/\sigma^{m/n} \quad (10)$$

$$t_B = C_B/\sigma^p \quad (11)$$

where C_D and C_B are defined by comparison with equations (3) and (8). Equating (10) and (11) gives

$$t_{D\text{-}B} = \frac{C_D}{(C_D/C_B)^{(m/n)/[(m/n)-p]}} \quad (12)$$

$$\sigma_{D\text{-}B} = (C_D/C_B)^{1/[(m/n)-p]} \quad (13)$$

EXPERIMENTAL INPUT

In order to determine how σ, a_0 and T control the ductile–brittle transition, the experimental values for the various parameters must be inserted into equation (9). The parameters will vary somewhat for the mode of loading, bending or tension, and on the thermal history of the material. Specific data will now be presented for single edge-notch tension tests under plane strain conditions for Marlex 6006 which was slowly cooled from the melt.

Figure 2 exhibits the creep behaviour of Marlex 6006 at 30°C. Up to about a strain of 10% the curves are fairly well described by equation (1). Analysis of *Figure 2* yields the following values of the parameters in equation (1): A (30°C) = 0.019 with $\varepsilon(\%)$, $m = 1.8$ and an average value of $n = 0.25$ (for stresses below 15 MPa). The parameter Q_D in equation (3) is $Q_D = 142\,000\ \text{J mol}^{-1}$ from Crissman

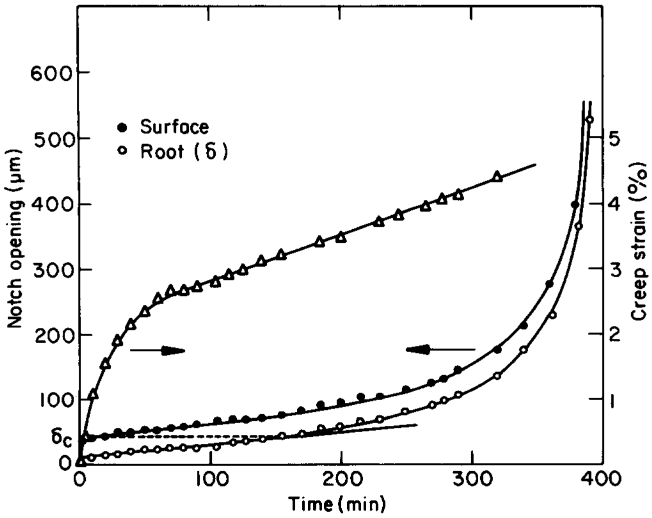

Figure 6 Crack opening displacement (circles) and creep strain (triangles) *versus* time: $a_0 = 246\ \mu\text{m}$, $\sigma = 10$ MPa, $T = 30$°C. From ref. 1

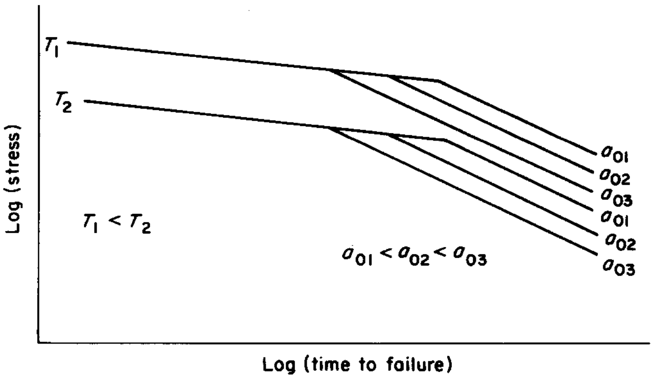

Figure 7 Schematic of equations (10) and (11)

and Zapas[1]. Consequently $D = 2 \times 10^{-18}$. With the above values and $\varepsilon_c = 10$, and for t_D with the units of minutes, from equation (3)

$$C_D = D\varepsilon_c^{1/n}\exp(Q_D/RT) = 2 \times 10^{-14}e^{142\,000/RT} \quad (14)$$

From *Figures 3* and *5*, $p = 5.0$ and $q = 1.9$. Measurements on the effect of temperature gave $Q_B = 100\,000\ \text{J mol}^{-1}$. Also $B = 2.3 \times 10^6$. The COD at which the δ vs. t curve becomes non-linear is $\delta_c = 27\ \mu\text{m}$. The ratio of the COD to the length of the damaged zone is $\alpha = 1/6$. Thus,

$$C_B = \frac{(a_0/6 + 27)e^{100\,000/RT}}{2.3 \times 10^6 \times a_0^{1.9}} \quad (15)$$

t_B is in minutes and a_0 is in micrometres.

The time $t_{D\text{-}B}$, at which the ductile–brittle transition occurs and $\sigma_{D\text{-}B}$ can now be calculated:

$$t_{D\text{-}B} = \frac{2 \times 10^{10} e^{5000/RT}(a_0/6 + 27)^{3.3}}{a_0^{6.2}} \quad (16)$$

$$\sigma_{D\text{-}B} = \left(\frac{4.6 \times 10^{-8} e^{45\,000/RT} a_0^{1.9}}{(a_0/6 + 27)}\right)^{0.45} \quad (17)$$

Schematic plots of (10) and (11) are shown in *Figure 7*. The slopes of the curves in the brittle region are greater than for the ductile region. These slopes are independent of temperature. The larger the defect the more the brittle regime is extended. The temperature dependence of the time to failure for a given stress is determined by Q_D for t_D and by Q_B for t_B. The temperature dependence of $t_{D\text{-}B}$ is much weaker since it is determined by $Q_D - (Q_D - Q_B)^{(m/n)/[(m/n)-p]}$. *Figure 7* is in qualitative agreement with the extensive experimental data that have been obtained from hydrostatic pressure tests on pipe as well as from other types of specimens[7,9].

BLUNTING EFFECT NEAR $t_{D\text{-}B}$

The above analysis is only valid if there is no interaction between the ductile failure process of general creep and the localized failure processes of slow crack growth. However, in the neighbourhood of the ductile–brittle transition there is a blunting effect on the notch by the creep process. *Figure 8* shows the blunting and broadening of the damaged zone as compared to the damaged zone produced at low stresses (*Figure 4*).

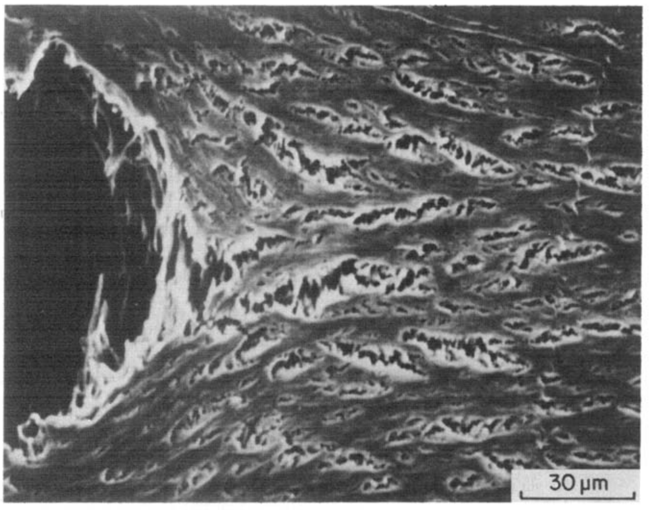

Figure 8 Microstructure of damaged zone at high stress showing blunting of the notch. Single edge-notched tension: $a_0 = 240\,\mu m$, $\sigma = 15$ MPa, $T = 30°C$, loading time = 83.7 min

Consequently the notch becomes less effective for a given value of a_0. This is a complex phenomenon whose analysis requires data on how the notch radius is changed by creep and the subsequent effect on the fracture process. There are experimental data by Iimura and Nishio[9], Szpak and Rice[10] and Bragaw[11] which demonstrate the effect of blunting. *Figure 9* shows the effect of blunting where the ductile region overshoots that time which would be predicted from the above analysis[10].

SUMMARY

Based on the creep equation, equation (1):

$$\varepsilon = A\sigma^m t^n$$

the time for ductile failure is given by equation (3):

$$t_D = \frac{D\varepsilon_c^{1/n}\exp(Q_D/RT)}{\sigma^{m/n}}$$

Based on the initial rate of damage from a notch, equation (7):

$$\dot{\delta}_0 = B\sigma^p a_0^q \exp(Q_B/RT)$$

the time for brittle failure is given by equation (8):

$$t_B = \frac{(\alpha a_0 + \delta_c)\exp(Q_B/RT)}{B\sigma^p a_0^q}$$

The time and stress for the ductile–brittle transition as a function of a_0 and T were obtained by equating equations (3) and (8).

The above theory does not take into account the blunting effect on the notch which occurs in the neighbourhood of the ductile–brittle transition. The numerical values of the parameters needed for specific calculations using the above equations have all been derived from creep and crack growth experiments and are given in this paper.

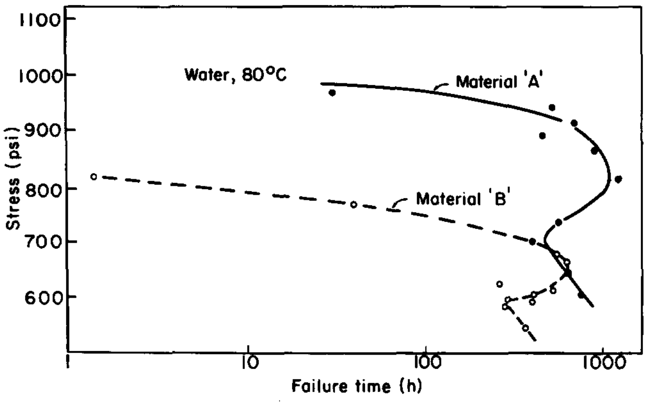

Figure 9 Hoop stress *versus* time to failure for gas pipes exposed to a hydrostatic pressure. From ref. 10

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REFERENCES

1 Lu, X. and Brown, N. *J. Mater. Sci.* 1986, **21**, 2423
2 Lu, X. and Brown, N. *J. Mater. Sci.* 1986, **21**, 4081
3 Crissman, J. M. and Zapas, L. J. *Polym. Eng. Sci.* 1979, **19**, 99
4 Zapas, L. J. and Crissman, J. M. *Polym. Eng. Sci.* 1979, **19**, 104
5 Hin, T. S. and Cherry, B. W. *Polymer* 1984, **25**, 727
6 Brown, N. in 'Yield Behavior of Polymers', (Eds. W. Brostow and R. D. Corneliusen), Hanser, New York, 1986, Chap. 6
7 Cooney, J. L. *J. Appl. Polym. Sci.* 1964, **8**, 1889
8 Lu, X., Bassani, J. L. and Brown, N. 'The *J*-Integral as a Fracture Criterion for Slow Crack Growth in Linear Polyethylene', Second Annual Report to Gas Research Institute, 1986
9 Iimura, S. and Nishio, N. 'Eighth Plastic Gas Pipe Symposium', Am. Gas. Assoc., Alexandria, VA, 1983, p. 34
10 Szpak, E., Jr, and Rice, F. G. 'Sixth Plastic Gas Pipe Symposium', Am. Gas Assoc., Alexandria, VA, 1978, p. 23
11 Bragaw, C. G. *ibid.*, 1978, p. 36