A fracture toughness study on low density and linear low density polyethylenes

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Fracture mechanics tests on two low density polyethylenes and two linear low density polyethylenes are described. The very low yield stresses give rise to crack blunting but at temperatures $<0°C$ crack growth occurs. The LDPE grades had rather low toughness but LLDPE gave much greater values and recourse to J methods was necessary. LLDPE materials would appear to have good prospects as tough, engineering plastics.

(Keywords: toughness; LDPE; LLDPE)

INTRODUCTION

Fracture mechanics is now well established for characterizing the toughness of polymers via the size and geometry independent parameters K_c, G_c and J_c[1]. Particular impetus to this work has been provided by the need to characterize high density polyethylenes used in pipe applications[2] and especially some medium density versions of these materials[3]. If linear elastic fracture mechanics (LEFM) is to be used then certain rather stringent size criteria have to be satisfied[2];

$$a,B,W-a>2.5\left(\frac{K_c}{\sigma_y}\right)^2 \tag{1}$$

where a is crack length, B is thickness, W specimen depth and σ_y is the yield stress. For high density polyethylene at 20°C, $K_c \sim 2\,\mathrm{MPa\,m^{\frac{1}{2}}}$ and $\sigma_y \sim 20\,\mathrm{MPa}$ giving a size parameter of about 8 mm which is fairly easy to achieve. The medium density materials have much higher toughness values; e.g. $K_c \sim 6\,\mathrm{MPa\,m^{\frac{1}{2}}}$ and $\sigma_y \sim 20\,\mathrm{MPa}$ so that the size is 225 mm. Specimens of this size cannot be made and recourse to the J_c method[3] has been necessary where the size parameter is much less;

$$a,B,W-a>25\left(\frac{J_c}{\sigma_y}\right) \tag{2}$$

which is 40 mm for a typical value of $J_c \sim 30\,\mathrm{kJ\,m^{-2}}$. Such specimens can be made and hence valid, plane strain, fracture toughness values obtained.

An even stronger challenge is provided by low density polyethylenes where $\sigma_y \sim 10\,\mathrm{MPa}$ and a considerable range of K_c values have been reported. For conventional low density PE materials K_c appears to be rather low ($\sim 1\,\mathrm{MPa\,m^{\frac{1}{2}}}$), but since there is little interest in these materials for engineering applications they have not been closely studied. For temperatures less than 0°C brittle fracture occurs and sizes of about 20 mm give valid LEFM data. However, transitions to very ductile behaviour occur at higher temperatures and the tests are no longer valid. The advent of linear low density polyethylene has changed the prospects for these materials, however, since they appear to have very high toughness values. Currently their use has been restricted mainly to film and other non-critical applications but their potential as very tough engineering materials seems considerable. With this in mind, therefore, a study is reported here in which four materials, two LDPE's and two LLDPE's are compared over a range of temperatures. The latter are very difficult to test in plane strain but the use of the J_c method has been explored here for this purpose.

THE CHARACTERIZING PARAMETERS OF FRACTURE MECHANICS

The linear fracture parameters K_c and G_c

The theory of linear elastic fracture mechanics (LEFM) deals with crack initiation occurring at nominal stresses that are well below the uniaxial yield stress of the material. Tests on pre-cracked specimens undergoing little plastic deformation are carried out to measure the fracture toughness, K_c, which characterises the elastic field around the crack tip. For single-edge notched specimens loaded monotonically, K_c is given by

$$K_c = Y(a/W)\sigma_c\sqrt{a} \tag{3}$$

where $Y(a/W)$ is a geometrical correction factor, σ_c is the gross applied stress and a is the initial crack length.

For single-edge notched bend specimens (SENB) with $S/W=4$, the geometrical correction factor is given by[4]

$$Y(a/W)=1.93-3.07\,(a/W) + 14.53\,(a/W)^2-25.11\,(a/W)^3+25.8\,(a/W)^4 \tag{4}$$

K_c is related to the energy per unit area of the fracture G_c by the relationship

$$K_c^2=\frac{E}{1-\nu^2}G_c \tag{5}$$

where E is the elastic modulus.

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Since fracture processes are controlled by the crack tip stresses and strains, and the states of triaxial stresses near the crack tip of a specimen are influenced greatly by specimen size, the fracture parameter K_c is therefore expected to vary with the size of specimen used. The material toughness is best characterized by its value under plane-strain conditions, K_{c1} and to achieve this state of stress the specimen dimensions must exceed some multiple of the plastic zone size, r_p. This limitation on the specimen size forms the basis of the minimum test-piece size requirements of the ASTME-399 standard for K_{c1} determination[4]; i.e. that

$$a, B, W-a > 2.5\left(\frac{K_{c1}}{\sigma_y}\right)^2$$

$$W > 2B$$

where B and W are the specimen thickness and width respectively and σ_y is the uniaxial yield stress.

Such limiting size requirements for the use of LEFM in determining K_{c1} in relatively brittle materials present no practical difficulties and K_{c1} determination can be carried out on a reasonably sized specimen[5]. However, characterizing the fracture toughness in tough materials would necessitate the use of much larger specimens which are difficult to manufacture hence an extension to LEFM has been provided by the development of ductile fracture mechanics to determine K_{c1} on smaller sized specimens than otherwise needed for LEFM.

The elastic–plastic parameter J_c

The restriction of small-scale yielding places a severe limitation on the application of LEFM; a restriction which effectively excludes lower strength materials and in particular polymers in which their poor thermal conductivity inhibits manufacturing thick sheets. As a result an elastic–plastic fracture parameter called the J-integral has been proposed and requires the determination of the value of J characterizing the initiation of crack extension, J_c.

The J-integral as originally defined as a path independent line integral for two dimensional problems can be expressed in terms of energy as[6]

$$J = -\frac{1}{B}\frac{dU}{da}\bigg|_{\text{at constant displacement}} \qquad (6)$$

where U is the potential energy of the loaded body (the area under the load–deflection curve). This energy definition of the J-integral was proposed as a fracture criteria for elastic–plastic behaviour of metals and extended the LEFM concepts to cases in which large scale plasticity is involved. Equation (6) was later expresses as[7,8]

$$J = \frac{\eta_e U_e}{Bb} + \frac{\eta_p U_p}{Bb} \qquad (7)$$

where U_e and U_p are elastic and plastic energy components of the total energy U, respectively, η_e and η_p are their corresponding elastic and plastic work factors and b is the uncracked ligament ($b = W - a$).

For three-point bend single-edge notched specimens having $a/W > 0.15$, $\eta_p = 2.0$.[7] Also when these specimens are tested with a span of $4W$ and have $0.4 < a/W < 0.6$, $\eta_e = 2.0$,[8] so that equation (7) reduces to

$$J = \frac{2U}{Bb} \qquad (8)$$

Equation (8) provides the basis for determining J_c using the multiple specimen 'R-curve' method.

For fracture to be characterized by J_c, a specimen must also meet certain size constraints in order to generate a plane-strain constraint along the crack front. To achieve this stress state all specimen dimensions must exceed some multiple of J_c/σ_y. According to ASTM[9], a valid J_c value may be determined when ever

$$B, b, W > 25\left(\frac{J_c}{\sigma_y}\right) \qquad (9a)$$

$$\omega = \frac{b}{J_c}\frac{dJ}{d(\Delta a)} > 1 \qquad (9b)$$

The condition given by the parameter ω, allows a J-field to dominate after a certain (small) amount of slow crack growth (Δa) and thus justifies the use of J after the onset of crack extension.

J_c can be directly related to the K_{c1} for linear elastic behaviour via the relationship

$$K_{c1}^2 = EJ_c/(1-\nu^2) \qquad (10)$$

so that $J_c = G_c$ for the LEFM case.

EXPERIMENTAL PROCEDURE

Materials

The test materials were supplied by BP Chemicals Limited in the form of 24 mm thick compression moulded sheets. The values of melt flow index (*MFI*) and density for various grades are listed in *Table 1*.

Yield stress

Tensile yield stress measurements were performed with dumbell shaped specimens (*Figure 1a*) using an Instron testing machine at a constant crosshead speed of 2 mm/min over the temperature range +20°C to −120°C. The load–time plots for each specimen were recorded and the yield stress σ_y calculated from the maximum load and the original cross-sectional area of the specimen. The variations of the tensile yield stress with temperature for LDPE and LLDPE are shown in *Figures 2a* and *2b* respectively.

Table 1 Materials used

Designation	Material type	Melt flow index (g/10 min)	Density (kg m^{-3})
A	Linear low density —film grade	0.80	920
B	Linear low density —film grade	1.0	924
C	Low density —general purpose	0.2	922
D	Low density	2.0	922

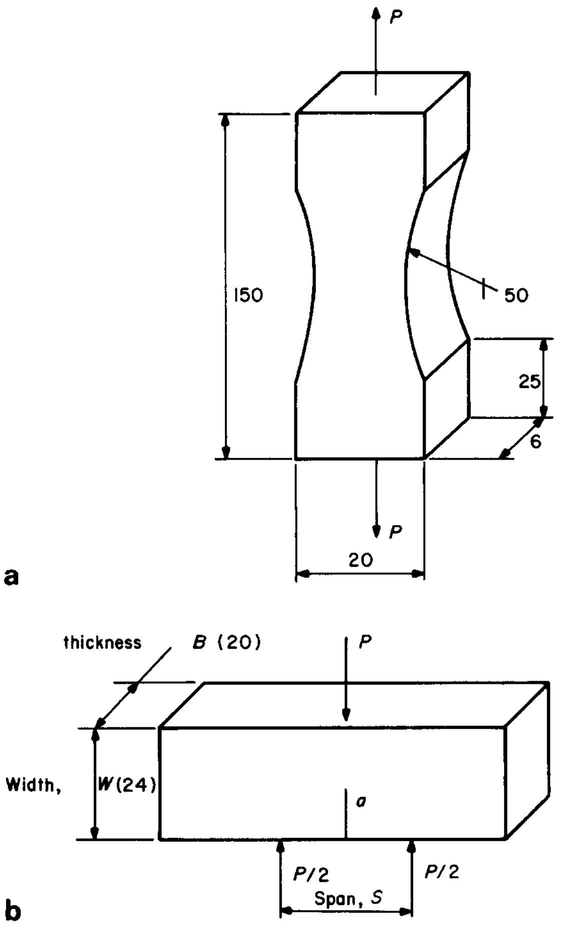

Figure 1 Specimen configurations (all dimensions in mm)

Fracture tests

The procedure used involves the measurement of load, load-line displacement and the crack extension, and from these measurements the J_c and K_c values may be calculated. All fracture tests were carried out on a single-edge notched bend specimens of dimensions $B=20$ mm, $W=24$ mm and length 150 mm (*Figure 1b*) loaded in three-point bending over a span of $4W$. Specimens were machine notched at room temperature using a single point fly cutter of tip radius $\sim 13\ \mu m$. Notches were introduced to a length greater than or equal to half the specimen width $W(a/W>0.5)$. The tests were performed on an Instron testing machine at a constant crosshead speed of 2 mm/min over the temperature range $+20°C$ to $-120°C$. The low temperature tests were carried out in an insulated box using liquid nitrogen vapour as the cooling medium. The temperature inside the box was controlled to an accuracy $\pm 1°C$ by a 'Eurotherm' Control Unit and a thermocouple located close to the crack tip.

The load-line displacement was measured with an LVDT attached to the loading nose and plots of load *versus* load-line displacement were recorded for each specimen on an X–Y recorder. Typical tests records for both grades of LDPE and one grade of LLDPE (A) at several test temperatures are shown in *Figures 3a* to *3c*.

FRACTURE TOUGHNESS (J_c) DETERMINATION

In accordance with the recommended procedure for establishing J_c outlined by ASTM[6], the multi-specimen R-curve method was used to characterize the ductile fracture toughness behaviour of LDPE and LLDPE at each test temperature. More specifically, a number of identical deeply cracked three-point bend specimens were loaded to various displacements producing different amounts of crack extension, Δa, and then unloaded. After unloading each specimen was broken open after immersion in liquid nitrogen so that the amount of crack extension could be measured.

The value of J for each specimen was determined from the area under its load *versus* load-line displacement curve and the relationship given by equation (8). The R-curve ($J-\Delta a$) was then constructed and J_c was taken to be the value of J where the R-curve intersected the blunting line as shown schematically in *Figure 4*.

Crack tip blunting

During the initial loading of a pre-cracked specimen crack tip blunting causes a stretched zone prior to material separation. The crack extension associated with this stretch zone Δa_b, can be approximated by assuming the stretch zone to be equal to half the crack opening displacement (*COD*), *i.e.*

$$\Delta a_b = \tfrac{1}{2}(COD) \qquad (11)$$

The *COD* is then related to J by the following relationship

$$J = m\sigma_y(COD) \qquad (12)$$

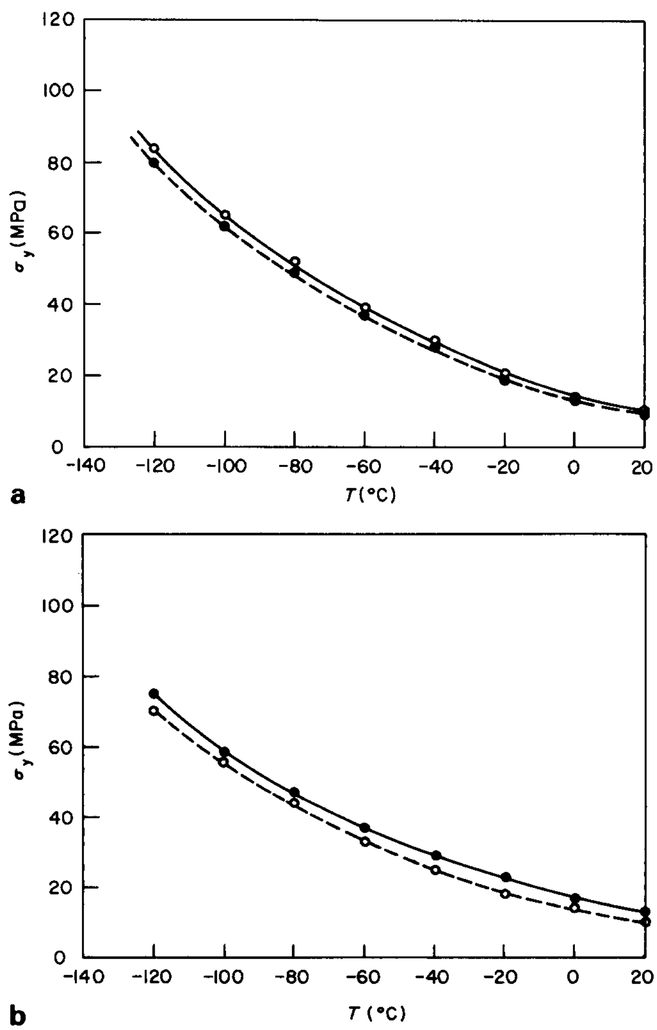

Figure 2 (a) Effect of temperature on the tensile yield strength of low density polyethylene (○) D, (●) C. (b) Effect of temperature on the tensile yield strength of linear low density polyethylene (●) B, (○) A

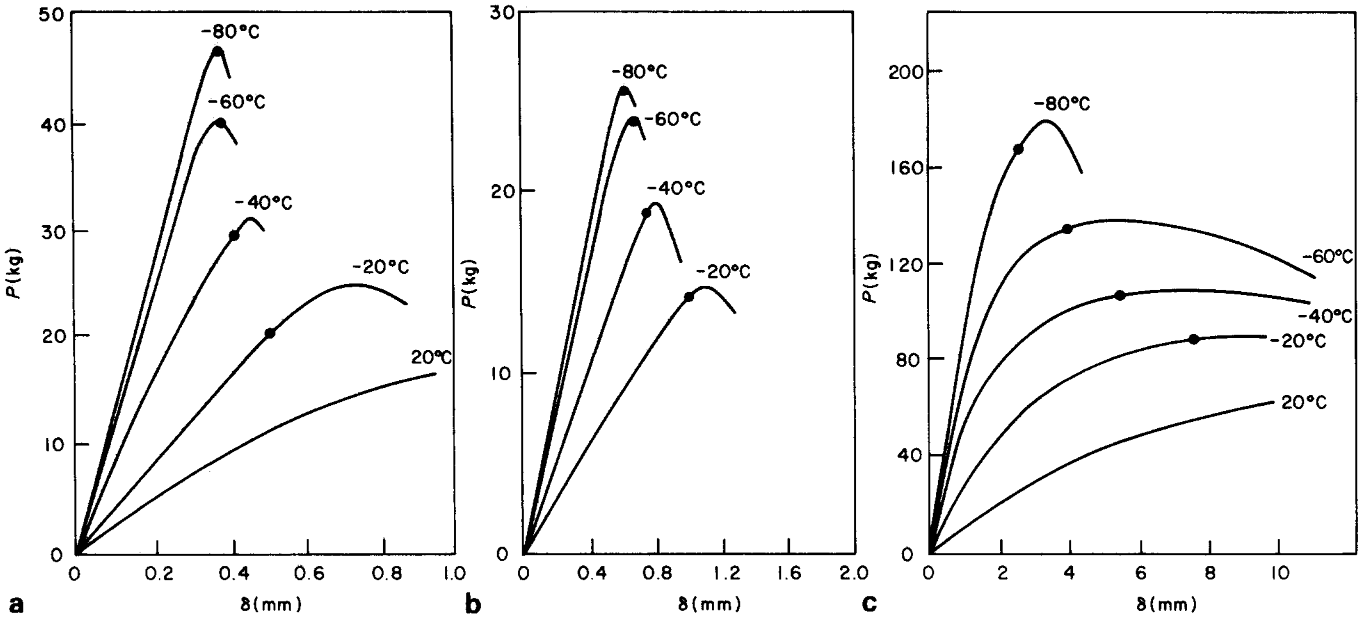

Figure 3 Load–displacement curves for SEN bend specimens of grades (a) C, (b) D and (c) A at various test temperatures (● initiation)

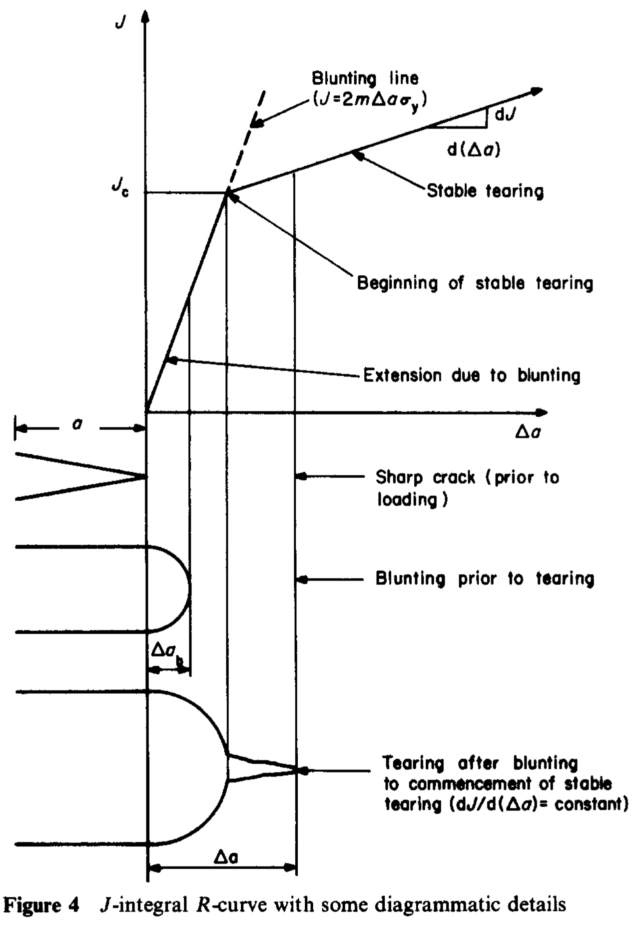

Figure 4 J-integral R-curve with some diagrammatic details

where m is the plastic constraint factor. Combining equations (11) and (12) results in a blunting line equation of

$$J = 2m\Delta a_b\sigma_y \tag{13}$$

In the standard J_c test procedures, the value of m is assumed to be unity. However, it has been reported recently that the value may depend on the material, the state of stress and the specimen size[10–12] and values of m in the range 1 to 3 are reported. The J_c values reported here are determined by using the value of m which best describes the experimental points.

Data analysis

In constructing an R-curve the following restrictions were observed:

$$\frac{\mathrm{d}J}{\mathrm{d}(\Delta a)} < 2m\sigma_y \tag{14a}$$

$$\Delta a < 0.06b \tag{14b}$$

Condition (14a) ensures that the slope of the R-curve is less than the slope of the blunting line and condition (14b) recommended by ASTM[6] ensures that the unloading due to the crack extension is sufficiently small so that the J-values remain path independent. However, there follows some experimental evidence to suggest that the allowable crack extension given by condition (14b) can be unnecessarily restrictive for LDPE and LLDPE.

RESULTS AND DISCUSSIONS

LDPE

Test records of load (P) *versus* loadline displacement (δ) for both grades of LDPE are shown in *Figures 3a* and *3b* for several test temperatures and exhibit a marked change in shape as the test temperature reduces from 20°C to −20°C. At −20°C and below the curves for both grades are similar in shape and are linear up to a certain load beyond which they departed from linearity but the load continued to rise until a well-defined maximum load was reached. In contrast the curves obtained at 20°C had broad peaks which were not well defined and the total displacements were significantly larger.

The comparable trends in the load–deflection curves for the two grades of LDPE revealed that, for temperatures below −20°C, C, with the lower MFI value, has a considerably greater maximum load and overall displacement thereby having a superior fracture resistance in comparison with D. It is significant to note that no instability condition was encountered in any of the specimens tested here over the whole temperature range and because of this attention was focused on determining the fracture initiation stage and its characterizing parameter, J_c, using the multi-specimen R-curve method.

Fracture surfaces obtained over the entire temperature range exhibited thumbnail features associated with the ductile tearing failure mode. The room- and low-temperature R-curves for both grades are illustrated in *Figures 5* and *6*. As indicated by the *Figures 5a* and *6a* all data points obtained at 20°C conform to a blunting line $J=2\Delta a\sigma_y$ and because of this the J_c value could not be determined. Furthermore during the specimen loading the crack tip was seen to blunt extensively with large amounts of plasticity in evidence. This eventually led to the plastic collapse of the specimen and the broad maximum that was seen on the load–deflection curves.

In the temperature range −20°C to −60°C, well defined R-curves were obtained for both grades and the J_c value at each test temperature was determined at the intersection of the R-curve and the blunting line $J=2\Delta a\sigma_y$. The initiation points are shown on the load–deflection curves in *Figures 3a* and *3b*. It is evident from the Figures that the initiation of the slow stable crack growth occurs prior to the attainment of the maximum load for specimens tested at −20°C and −40°C. For specimens tested at −60°C and below initiation coincided with the maximum load on the P–δ curves.

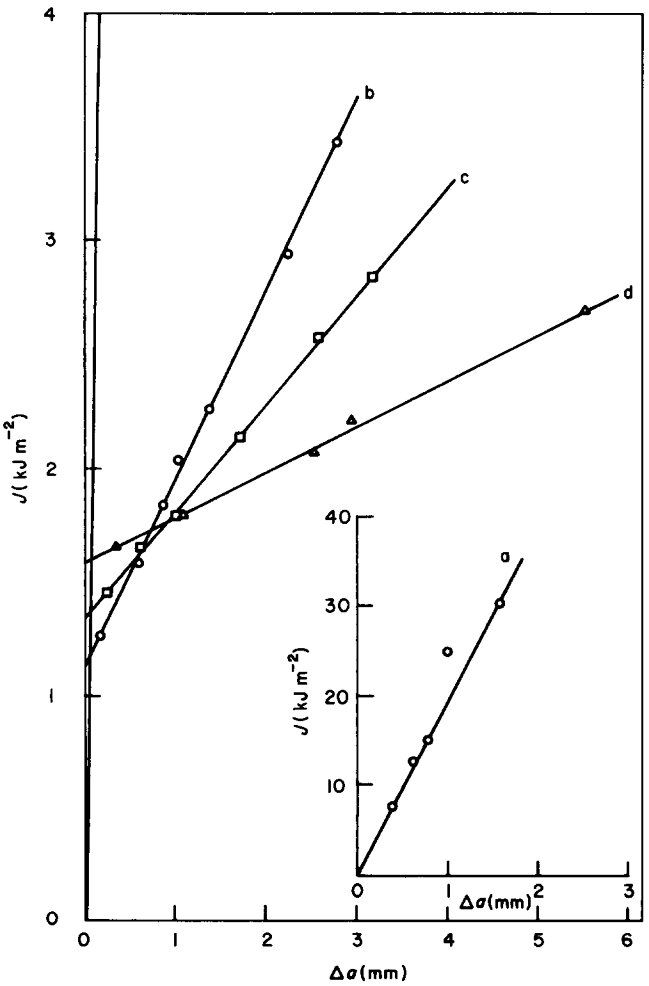

Figure 5 J–Δa curves for C at various test temperatures; (a) 20°C, (b) −20°C, (c) −40°C, (d) −60°C

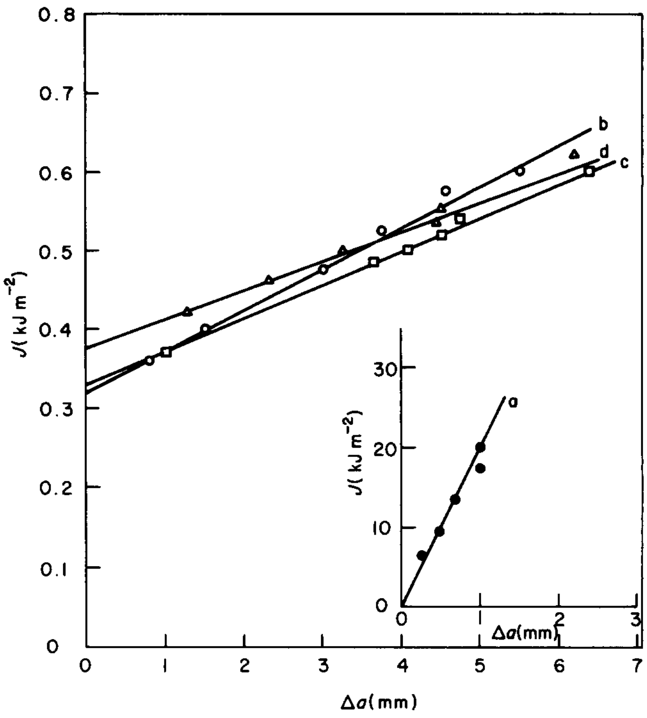

Figure 6 J–Δa curves for D at four temperatures; (a) 20°C, (b) −20°C, (c) −40°C, (d) −60°C

At temperatures below −60°C the J_c values were calculated at the maximum load, P_{max}, via the relationship given by equation (15) rather than recourse to the R-curve method:

$$J=\frac{2U_{max}}{Bb} \tag{15}$$

This was done because of difficulty in controlling Δa when $dJ/d\Delta a$ is small. The use of equation (15) was justified since no evidence of ductile tearing was visible on the fracture surfaces of those specimens which were unloaded just prior to the attainment of the maximum load.

The effect of temperature on J_c, $K_c(J_c)$ and $dJ/d(\Delta a)$ values for both grades of LDPE is shown in *Figures 7* and *8* (the mean value of J_c is presented for specimens tested below −60°C). $K_c(J_c)$ values were evaluated from equation (10) and $E/(1-\nu^2)$ was calculated from the measurements of the initial slope of the load–deflection curve and substituted into the appropriate elastic compliance expression[13].

The Figures show that J_c and K_c values for both grades increase with decreasing temperature. A comparison of *Figures 7* and *8* indicates the influence of *MFI* on the fracture parameters. Evidently of the two grades C with the lower *MFI* exhibits a superior J_c, K_c and $dJ/d(\Delta a)$ to that of D having a higher *MFI* value. Indeed the data suggest that a ten-fold drop in *MFI* results in an approximately ten-fold increase in $dJ/d(\Delta a)$ and approximately tripled the fracture initiation values J_c.

The parameter $\omega=b/J_c dJ/d(\Delta a)$ was also calculated for both grades and values in the range of 1.3 to 7.5 for C and 1.0 to 2.0 for D were obtained in the temperature range −60°C to −20°C. These values suggested that for the specimen sizes used here the singularity is dominant at the crack tip during the J-controlled fracture.

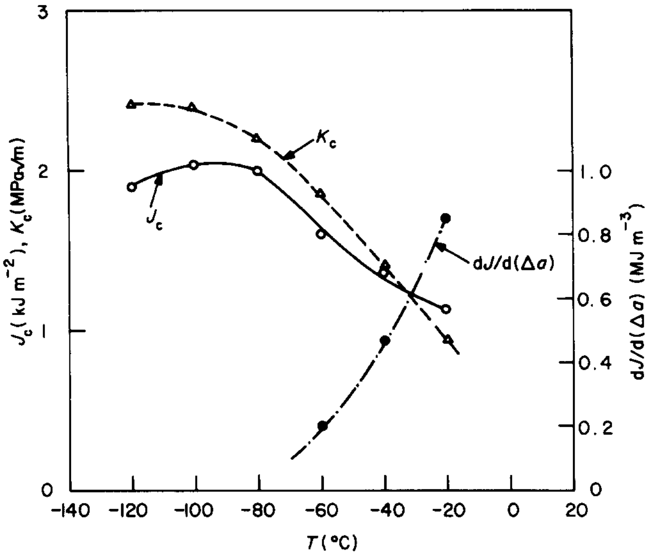

Figure 7 Effect of temperature on J_c, K_c and $dJ/d(\Delta a)$ for C (K_c calculated from equation (10))

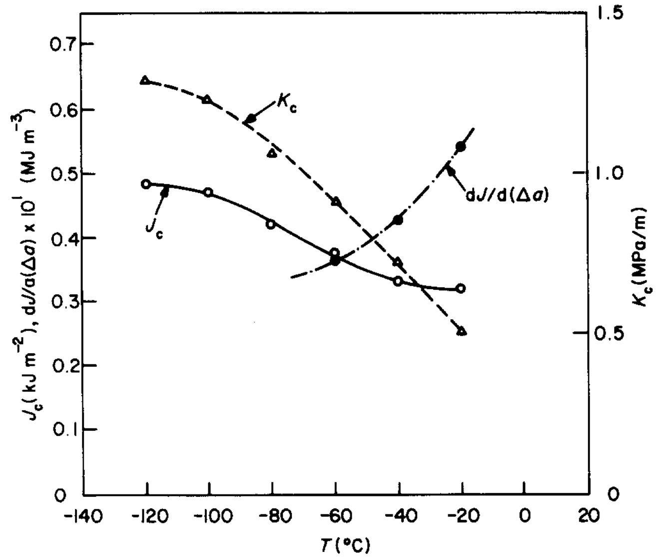

Figure 8 Effect of temperature on J_c, K_c and $dJ/d(\Delta a)$ for D (K_c calculated from equation (10))

It should be noted that although the J method was used for these materials the LEFM maximum stress expression (equation (1)) could have been employed since the critical LEFM size $2.5\,(K_c/\sigma_y)^2$ varied from 11 mm at 0°C to 2 mm at -120°C for the tougher material (C) and even small values for D. K_c values calculated via this route agreed exactly with those given here since $\eta_e = 2$ for those specimens as stated previously.

LLDPE

As shown in *Figure 3c*, the early stages of deformation for LLDPE are elastic as reflected by the linearity of the load–deflection curves. As loading continued the crack tip was seen to blunt before moving through the rest of the specimen. The load continued to rise and eventually reached a maximum and decreased thereafter. The peak was broad at 20°C and not well defined but as the temperature was reduced it became progressively sharper and quite distinct. Comparison of *Figures 3a, b* and *c* illustrates the difference in the shape of the P–δ curves for LDPE and LLDPE. Although at room temperature the P–δ curves are similar at -20°C and temperatures below LLDPE exhibits a considerably greater total displacement coupled with a greater maximum load, thereby accounting for its superior fracture resistance in comparison to LDPE. The trends in load–deflection curves for the two grades of LLDPE indicated a slightly greater maximum load and total displacement for A than B. Fracture surface examinations of LLDPE specimens also revealed crack extension with the thumbnail profile associated with the ductile tearing failure made. No instability conditions were encountered in any of the specimens tested here.

The $J-\Delta a$ curves for both grades of LLDPE at various test temperatures are illustrated in *Figures 9* and *10*. As in the case for LDPE, the large crack tip blunting and fully yielded ligament did not allow the crack to extend at room temperature and consequently the J_c value could not be determined. The lines drawn in *Figures 9a* and *10a* represent a slope of $J/2\Delta a\sigma_y = 3$ (i.e. $m = 3$). At -20°C and temperatures below, well defined R-curves were obtained so that J_c values could be determined. It is clear from the figures, however, that the conventional blunting line with $m = 1$ does not describe the data for the whole temperature range. The value of m is apparently affected by the test temperature as shown in *Figures 11* and *12* which show that m decreases from about 3.0 at 20°C to 1.0 at -80°C and below.

The effect of temperature on J_c, and $dJ/(\Delta a)$ values are also shown in *Figures 11* and *12* for A and B respectively. Comparison of the Figures suggested that of the two grades, A, having a lower *MFI* value, has a better fracture properties than B, particularly at temperatures below -60°C. Both grades exhibited a transition in their $dJ/d(\Delta a)$ value at -60°C. The ω factor for LLDPE was in the range of 6 to 18 for A and 3.5 to 16 for B hence satisfying the J-control crack growth criterion. It should also be noted that the LEFM size criterion cannot be met in these materials except at very low temperatures but that the J-criteria is met for temperatures up to -80°C. For this reason the values at -60°C, -40°C and -20°C cannot be taken as valid and presumably the rise in m is a reflection of this change in the stress state.

CONCLUSIONS

There are several very striking features of the data presented here. In the first case the transition from cracking to a continuous crack tip blunting process was very marked in these materials. Presumably larger specimens would give crack growth since this large effect is due to the lack of constraint in the specimens and the very low yield stresses. When cracking does occur, however, the LDPE materials show rather low toughness values being in the range of K_c values of 1 to 2.5 and 0.2 to 0.6 MPa m$^{\frac{1}{2}}$ which would be similar to many brittle amorphous polymers. Their greater ductility is, therefore, a consequence of low σ_y values but as common experience shows once cracking occurs it is very brittle.

The LLDPE materials have qualitatively similar behaviour but the K_c values are extremely high being around 7 MPa m$^{\frac{1}{2}}$ with J_c and $dJ/d\Delta a$ being an order of magnitude or more higher. Toughness values of this

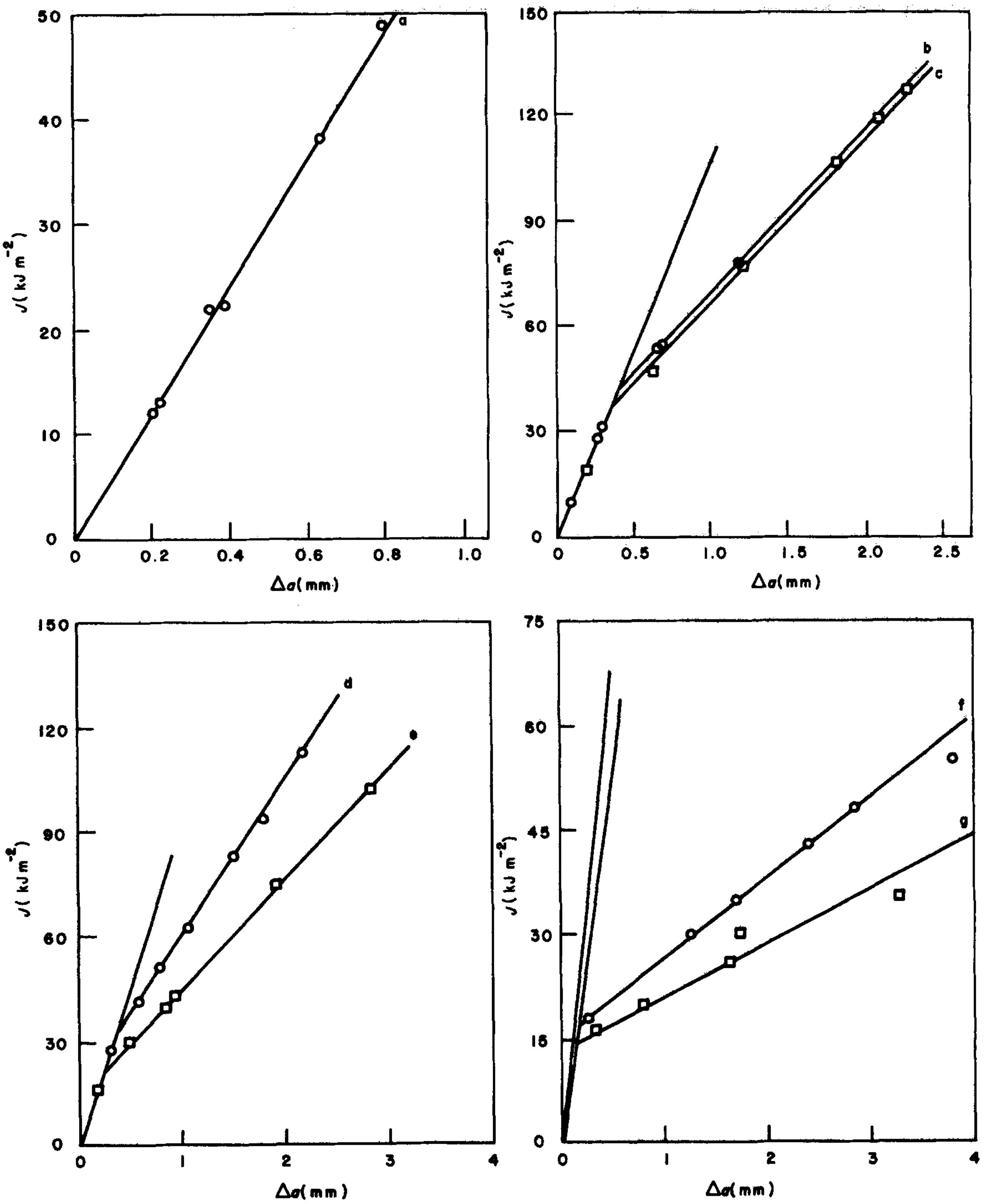

Figure 9 *J*–Δ*a* curves for A at various test temperatures; (a) 20°C, (b) −20°C, (c) −40°C, (d) −60°C, (e) −80°C, (f) −100°C and (g) −120°C

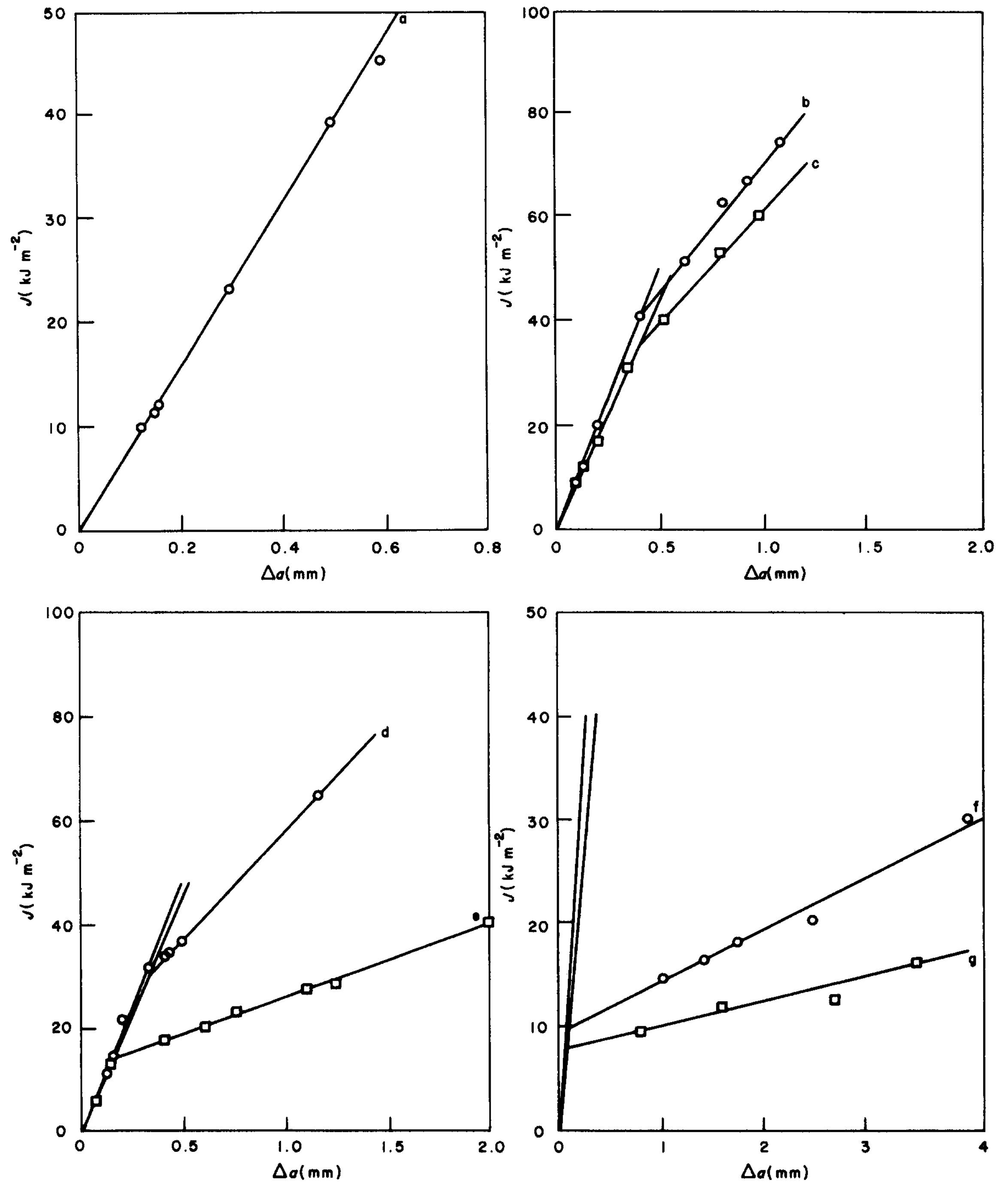

Figure 10 J–Δa curves for B at various test temperatures; (a) 20°C, (b) −20°C, (c) −40°C, (d) −60°C, (e) −80°C, (f) −100°C and (g) −120°C

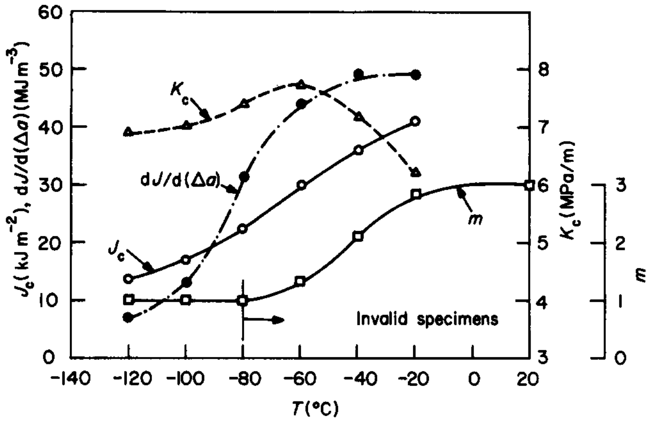

Figure 11 Effect of temperature on J_c, K_c, $dJ/d(\Delta a)$ and m for A (K_c calculated from equation (10))

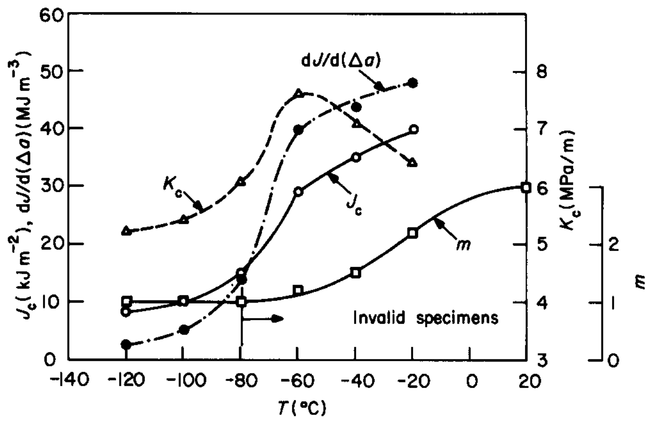

Figure 12 Effect of temperature on J_c, K_c, $dJ/d(\Delta a)$ and m for B (K_c calculated from equation (10))

magnitude have been reported for a MDPE[3] and for toughened nylon[14]; both semi-crystalline polymers. Presumably this difference arises in some way from the lack of short chain branching in these materials but they are clearly greatly different from HDPE's where K_c values of 1 to 2 MPa m$^{\frac{1}{2}}$ are more usual[2]. The exploration of these materials as engineering plastics would appear to be a good prospect.

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NOMENCLATURE

a	Crack length
Δa	Crack extension
Δa_b	Crack extension due to blunting
B	Specimen thickness
b	Length of uncracked ligament
E	Young's modulus
J	Value of J-integral
J_c	Value of J at the onset of crack extension
J_{max}	Value of J at the maximum load
$dJ/d(\Delta a)$	Rate of change of J with crack growth
K	Stress intensity factor
K_c	Critical value of K
K_{c1}	Critical value of K in plane strain
m	Yield stress constraint factor
P	Load
r_p	Plastic zone size
S	Span
T	Temperature
U	Deformation energy (area under the load–deflection curve)
U_e	Elastic strain energy
U_p	Work done in plastic deformation
U_{max}	Value of U at the maximum load
δ	Displacement
W	Specimen width
Y	Finite width correction factor
ν	Poisson's ratio
η_e, η_p	Constants for elastic and plastic work terms, respectively
σ	Stress
σ_c	Stress at initiation point
σ_y	Yield stress
ω	J-controlled crack growth parameter