JOURNAL OF MATERIALS SCIENCE 29 (1994) 4545–4553

The work of fracture in semiductile polymers

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The work to fracture in tension double-notched samples of some semiductile polymeric materials (rigid PVC, Orgalloy and Ultranyl) has been measured as a function of the ligament length. It was established that the work of fracture was proportional to the extent of the plastic zone that developed in the ligament area during crack propagation. It is proposed that the total energy density is made up of two terms, one distributed all over the plastic zone and the other localized in the vicinity of the fracture path. It is then shown that a linear relationship exists between the specific work of fracture and the ligament size, provided the height of the plastic zone linearly depends on the ligament length. The linear extrapolation of the specific work of fracture to nil ligament, yields a value that coincides with J_{1C} and therefore can be treated as a critical parameter. It is also shown that, at large ligaments, the dependence of the specific work of fracture on the ligament length reflects the post-yield behaviour of the material and it is influenced by the tendency of the height of the plastic zone to level off. Consequently, no specific meaning can be given to quantities obtained, according to the essential work of fracture theory, in the large ligament region, e.g. ligaments larger than three to five times the sample thickness.

1. Introduction

The progressive displacement of traditional materials by polymers, even for structural applications, renders the evaluation of their fracture resistance an exacting task. There are several methods to appraise the fracture resistance of rigid materials, all based on defining proper quantities whose value progressively rises as the severity of the external loading conditions increases. Critical values of these parameters (which depend on a number of variables such as temperature, strain rate or environmental interactions) are postulated to exist, so that crack propagation can occur when the actual value of such quantities exceeds their corresponding critical values. Particularly useful, from an engineering point of view, is the approach based on analysing the stress level around defects. According to Irwin [1] the stress level around an elastic crack is uniquely determined by the parameter K (stress intensity factor) which depends on the stress remotely applied, σ_0, to the cracked body (whose geometry is described by the parameter Y) and on the size of the crack, a, according to

$$K = Y\sigma_0 a^{1/2} \quad (1)$$

As the remote stress, σ_0, increases so does K. Irwin's model admits that, under specified test conditions, a critical K value, K_C, exists for all materials and that propagation occurs when $K > K_C$. The applicability of this model to small-scale yielding, compared with the size of the crack, a, can be gained if a fictitious crack length, a_{fic} [2] is used which incorporates a portion of the yielded region at the crack tip (well-contained plasticity). In plane stress

$$a_{fic} = a + (1/\pi)(K/\sigma_y)^2 \quad (2)$$

where σ_y is yield stress of the material.

This fracture criterion, very straightforward to use, is inapplicable when plasticity is not contained in a small region compared to the crack size (there are cases in which size requirements can dictate the use of specimens of hundreds of millimetres thick). Although the K-criterion is widely utilized in mechanical engineering, it is not very useful to materials scientists because it does not yield any basic material's parameter; the unusual dimensions of K, $N m^{-3/2}$, reveal its composite nature.

An approach that does not suffer from these limitations (it does not make any assumption on the nature of the stress–strain curve provided the stress steadily increases with the strain) is based on thermodynamic considerations. Griffith [3] considered an infinite sheet, containing a central crack and remotely loaded in tension, for which an extension of the original crack brings about a decrease of the potential energy of the system. If the energy thus made available is greater than the energy required to generate the additional free surface originated by the enlargement of the original crack, then propagation can take place. At the onset of crack growth we have, per unit thickness

$$G_C = -\delta U/\delta a = 2\gamma \quad (3)$$

where U is the strain energy, G_C is the critical strain energy release rate, and γ is the surface energy of the material. It was soon recognized that, for most materials, γ is much too small to account for the observed values of G_C; this discrepancy was eliminated by

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incorporating in γ the work done to produce the plastic zone that surrounds the crack tip [4]. In the case of elastic bodies, the stress at failure, in plane stress, is given by

$$\sigma_f^2 = 2E\gamma_{app}/\pi a \quad (4)$$

This relationship is important because it incorporates an engineering quantity (the stress), a material parameter (the apparent surface energy), and a geometrical term (the crack length) in a form that identifies the critical conditions. Being based on a linear stress–strain relationship, it suffers from the same limitations as the K approach. The J-integral concept set out by Rice [5] for elastic–plastic materials is also based on an energy balance and is presently widely utilized. For the sake of simplicity, J can be defined, similarly to Equation 3, as

$$J = -\delta U/\delta a \quad (5)$$

where U is the strain energy, irrespective of whether or not the stress–strain relationship is linear. The main difference between elastic and inelastic materials during crack propagation is that, for the former, the potential energy stored in the region adjacent to the crack path is reversibly released (and used up to drive the crack through) whereas, in the latter, the released energy is decreased by the amount required to deform the material plastically. If the difference between the input and output energies in the strained zones becomes important, insufficient energy remains to sustain the growth of the crack: fracture then propagates in a stable manner or even comes to rest. At the onset of crack propagation no volume elements are yet unloaded and the similarity between Equations 3 and 5 suggests defining the critical conditions as

$$J_C = -(\delta U/\delta a)_C \quad (6)$$

The problem with ductile materials is to identify the transition between subcritical and critical growth. The subcritical growth is associated with the development of an initial plastic zone which blunts the crack tip to a radius equivalent to half the CTOD. According to the standard procedure ASTM E-813, the critical value for J is thus given as the intercept of the blunting line $J = 2\sigma_y\Delta a$ and the propagation J versus Δa line [6]. This procedure is still at issue, particularly for polymers for which a number of irreversible processes can take place in the process zone, e.g. crazing or cavitation [7]. Besides this, experimental difficulties inherent in the J-integral method arise from the necessity to identify small movements of the fracture front and to stop propagation after starter cracks are caused to move. Under many experimental conditions, the method is consequently inapplicable (e.g. high-rate loadings or harsh environments in which samples are to be confined).

In the thermodynamic approach, the fracture resistance of a material is related to the energy required to create the new surfaces. This parameter scales with the surface and incorporates the energy dissipated in the plastic zone at the crack tip which, in turn, scales with the volume. The two energy terms can be combined if a linear term, h, is introduced that scales with the size of the plastic zone

$$\gamma_{app} = \gamma_{surf} + \gamma_{plast}/h \quad (7)$$

It is, therefore, clear that the size of the plastically deformed region, somehow measured by h, is of paramount importance, because γ_{plast} outweights γ_{surf}. The problem of the shape of the plastic zone is specifically addressed in the model proposed by Broberg [8–10] in which the *total* work of propagation, U_{TOT}, is considered, in contrast with the G and J methods which are based on the *incremental* quantity $\delta U/\delta a$. The model is based on the idea that the yielded zone surrounding the crack tip can be divided in two parts; an inner and an outer region. The energy required to make the crack move is also partitioned in two components: one, absorbed in the inner region close to the crack trajectory, was termed the essential work of fracture, U_E; the other, dissipated in the outer region that surrounds the inner one, was termed the non-essential work of fracture, U_{NE}. The model (EWF in short) is embodied in the simple equation $U_{TOT} = ltu_E + \beta l^2 t u_{NE}$. By referring all terms to the unit surface we have

$$u_S = u_E + \beta l u_{NE} \quad (8)$$

where l and t are the ligament size and the sample thickness, β is a shape factor of the outer plastic region, u_S ($= U_{TOT}/lt$), u_E, u_{NE} are the total specific, specific essential and specific non-essential works, respectively.

Two deformation regimes are identified depending on the size of the ligament relative to the sample thickness. At large ligaments, plane stress conditions prevail in the ligament and, according to Equation 8, a linear dependence between u_S and l is expected and u_E can be found by extrapolating to $l = 0$. In order for plane stress conditions to prevail, the requirement $l \geqslant 3$–$5t$ must be fulfilled; it is further required to have the ligament region fully yielded before the crack starts propagating (i.e. on load versus displacement plots the maximum should occur before propagation). On decreasing l, plane strain conditions progressively set in and the work of propagation consequently decreases. Because the ligament is now in a mixed-mode regime, the linearity breaks and the extrapolation of u_S data, in the small ligament interval, yields a value of u_E smaller than that obtained from large ligament data. To this value the meaning of a critical parameter, for the particular thickness being examined, can be given. Equation 8 was first used to measure the specific essential work of fracture for ductile tearing of thin sheet metals [11–16]. The same technique was subsequently extended to measure u_E for a range of ductile polymers in plane stress conditions using DENT geometry [17–21]. There are advantages associated with this model: the relevant geometric parameter is the ligament not the crack advancement and the energetic term is the total work of fracture. Both quantities are readily accessed under all test conditions. Two important drawbacks are (1) the relationship between u_S and l is non-linear over the entire ligament range, (2) Equation 8 does not yield any immediate definition of the critical conditions, (3)

the transition from mixed-mode to plane stress is ill-defined on u_S/l plots. These aspects are considered in this paper.

2. Experimental procedure

Three semiductile polymers were tested: unplasticized PVC (6 and 3 mm thick), Orgalloy 6000 (6.5 and 2 mm thick) and Ultranyl KR 4510 (9 and 4 mm thick); all but PVC were obtained from the VAMAS (Versailles Advanced Materials and Standards) project.

Orgalloy is a mixture of nylon-6 and polypropylene (40/60 by weight). Nylon-6 is the matrix in which polypropylene domains are dispersed. Ultranyl is a blend of nylon-66 (as the matrix) and poly(phenylene ether). In both materials the morphology is stabilized by block copolymers.

The work of fracture was measured in uniaxial tension using double-edge-notched testpieces (DENT) at a displacement rate of 10 mm min^{-1}. Testpieces were 150 mm long and 25–75 mm width. Sample thicknesses were 6 mm for PVC, 2 mm for Orgalloy and 4 mm for Ultranyl.

The J-integral was measured in three-point bending ($S/W = 4$) at the same displacement rate. Sample thicknesses were 6 mm for PVC, 6.5 mm for Orgalloy and 9 mm for Ultranyl.

The extent of the plastic zone was evaluated, after sample failure, by means of a TV camera equipped with a close-up lens. Image analysis was carried out by an image processing software. An example of the extent of the plastically damaged zone is shown in Fig. 1. All measurements were carried out at room temperature (23 °C).

3. Results and discussion

Let us consider the sample in Fig. 2, loaded in tension up to failure that occurs by propagation of the two opposite cracks (for fully ductile materials, deformation rather proceeds by stretching the ligament) and forming a yielded area confined in the ligament region.

We can split the total work of fracture, U_{TOT}, in two terms related to the formation of new surface and to

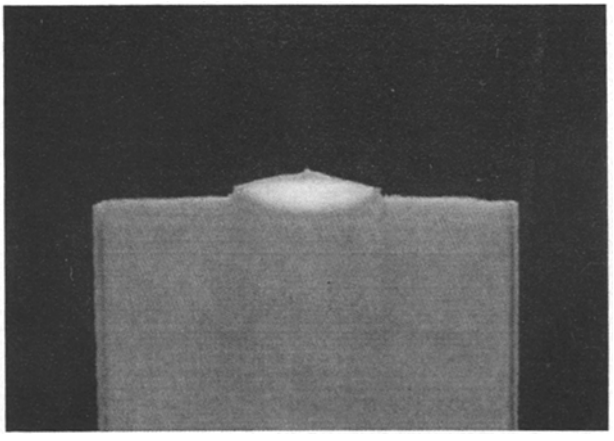

Figure 1 Plastic zone developed around the ligament in PVC 6 mm thick.

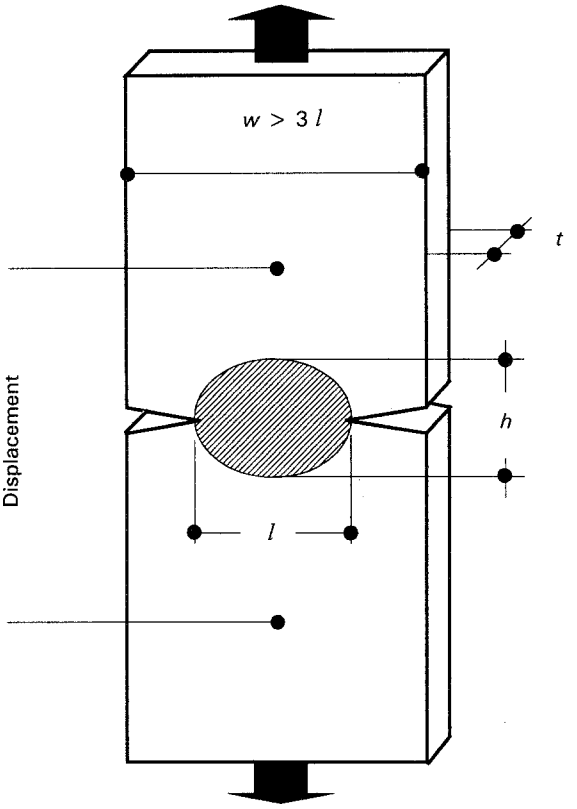

Figure 2 Development of plastic zone in a DENT specimen.

the development of the plastically strained region, of volume V_p, that surrounds the crack path. By taking into account that the surface term is negligible we can write the simple relationship

$$U_{TOT} = \int_{V_p} \Phi_P dV_P \quad (9)$$

where Φ_P is the energy density function, i.e. the work to be made to create a plastic zone of unit volume.

In a plastic body, the energy density within a strained region is given by the product of the average flow stress and the deformation of the material: $\Phi_P \simeq \sigma_{flow}\varepsilon_P$. Although we can equate the plastic flow stress, σ_{flow}, to the yield stress and take it as constant, it is unrealistic to also consider the plastic deformation, ε_P, as a constant over the entire plastic volume that develops in the ligament region. By measuring the deformation of the rubber particles within the plastic zone in some toughened nylons, it has been very recently shown [22] that the plastic deformation within the plastic zone around a crack rapidly increases on approaching the fracture path. If we approximate the real dependence of ε_P on d (distance from the fracture surface) with a step function, Fig. 3 top, it is possible to decompose the energy density function, Φ_P, into two terms; $\bar{\phi}$, constant over the whole plastic region and ϕ^*, which can be considered as an excess plastic energy density, restricted to the vicinity of the fracture path, Fig. 3 bottom. If the thickness of the region in which the excess plastic energy density is confined, h^*, does not change with l, we can rewrite Equation 9 as

$$U_{TOT} = \int(\phi^* + \bar{\phi})dV = \phi^* h^* l t + \bar{\phi} V_P \quad (10)$$

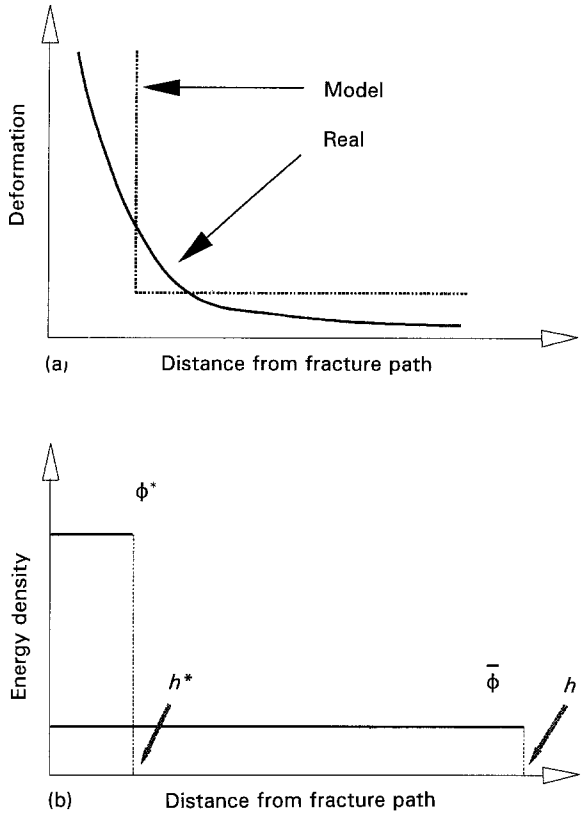

Figure 3 Schematic illustration of the distribution of (a) deformation and (b) energy density within the plastic zone.

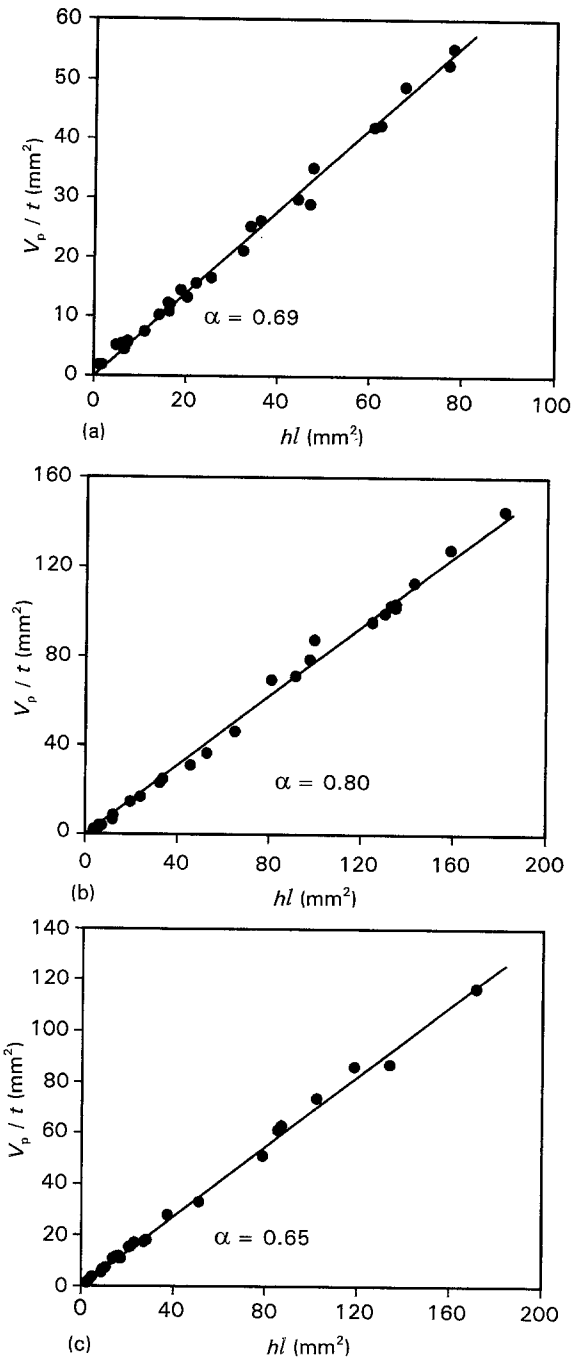

Figure 4 Evaluation of the shape factor α (Equation 11) for (a) Ultranyl, (b) PVC, and (c) Orgalloy.

It is experimentally difficult to determine the volume of the plastic region. In some cases, deformations beyond the yield point cause the nucleation and growth of defects, such as crazes or microvoids, which trace the yielded region by altering the local optical properties (the well-known stress-whitening). All the materials utilized in this work show a marked whitening and the transition from whitened to undamaged zones is reasonably sharp. By means of a high-resolution TV camera and an image processor, the volume of the damaged areas was measured for samples of different ligaments.

In principle, the plastic volume can be written as

$$V_P = \alpha(l, t)h(l, t)lt \qquad (11)$$

where $\alpha(l, t)$ is a shape factor (for instance, for an ellipse $\alpha = \pi/4$) and $h(l, t)$ is the height of the plastic region; both parameters possibly depends on l and t. It is possible to evaluate the influence of l on α by plotting the ratio V_P/t as a function of hl, Fig. 4. The linear dependence observed reveals that, for all materials and ligaments examined, α was substantially independent of l. Measurements carried out on samples of different thickness did not show any major influence of thickness on α; this aspect will not be further examined in this paper.

The issue is now to clarify the dependence of h on l. As shown in Figs 5–7, the height of the plastic zone increases on enlarging the ligament; two regions can be identified in the h versus l plots. At small ligaments, when the crack tips are close to each other, h almost linearly varies with l. Above a certain value, the dependence becomes loose and seemingly a plateau value would be eventually attained at very large ligaments. To explain this effect, two deformation regimes can be figured out. When the inter-crack distance is small, the stress fields at the crack tip interact and all the ligament is in a mixed-mode state of tension, i.e. neither full plane stress nor full plane strain. The size of the plastic zone in front of a crack is, in plane stress, approximately equal to $(K/\sigma_y\pi^{1/2})^2$, which for a polymer (PVC) with $K_{1C} = 7 \times 10^6$ (K_{1C} instead of K_{IC} because we are not referring to plane strain conditions) and $\sigma_y = 55$ MPa corresponds approximately to 5 mm. If the actual ligament is less than twice this value, the plastic zone cannot fully develop; consequently, h rapidly increases with l because the plastic constraint rapidly vanishes. The largest ligaments at which the linear relationship between h and l drops

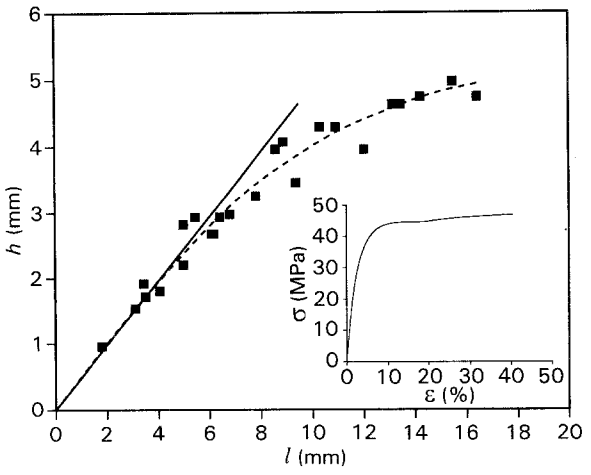

Figure 5 Height of the plastic zone as a function of ligament length for Ultranyl 4 mm thick. Inset: stress–strain curve.

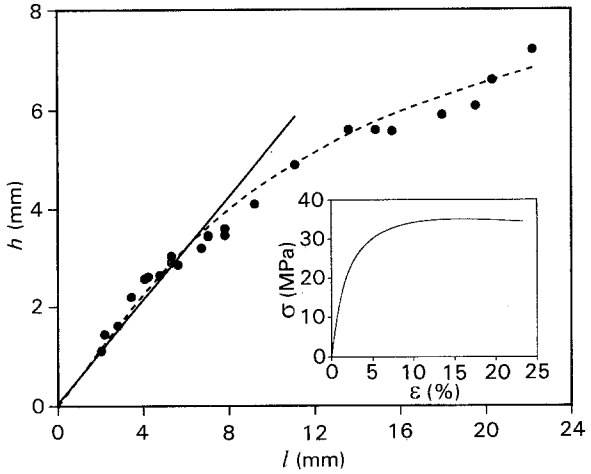

Figure 6 Height of the plastic zone as a function of ligament length for Orgalloy 2 mm thick. Inset: stress–strain curve.

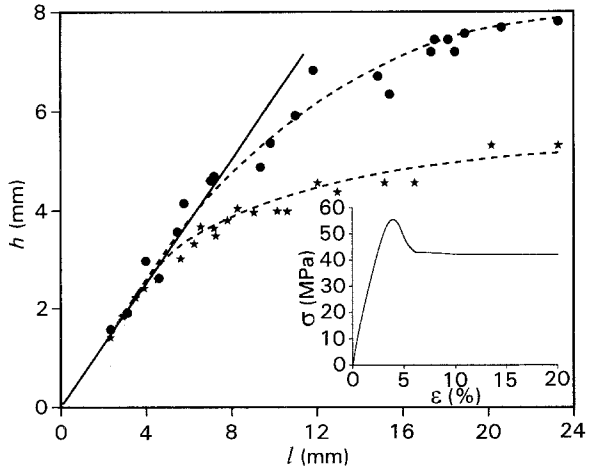

Figure 7 Height of the plastic zone as a function of ligament length for PVC (★) 3 mm and (●) 6 mm thick. Inset: stress–strain curve.

were about 8, 10, 8 and 8 for Ultranyl (4 mm), PVC (6 mm), PVC (3 mm) and Orgalloy (2 mm), respectively.

At large ligaments the resisting section can be seen to be made of three segments; the two terminals are related to the crack tips and the central one experiences stress conditions similar to those sensed in a simple tensile test. If we neglect the stress raising effect of the lateral cracks, the central portion will yield, due to the local increase of stress originated by the decrease in cross-section ($\sigma = \sigma_0 W/l$). The ligament therefore necks down and the geometry of the neck will depend on the level of strain softening. If the material strain-hardens (e.g. metals), plasticity can spread out, particularly at large ligaments when the stress intensification effect, originated by the change in cross-section, is locally counteracted by the increase of yield strength; for most polymers strain softening is conspicuous and occurs soon after yield, and plasticity consequently localizes. The dependence of h on l should therefore somehow reflect the yielding behaviour of the material. The development of the plastic zone can be schematically depicted as in Fig. 8. It is of interest to note that for PVC, a material that shows a considerable strain softening (see the stress–strain curve in the inset in Fig. 7) the tendency of the height of the plastic zone to level off is more evident than for Orgalloy and Ultranyl, which do not exhibit a stress drop at yield (see the insets in Figs 6 and 7). It was also observed that the plateau values scale with the thickness of the samples as shown, as an example, in Fig. 7 for PVC. The upper limit of the height of the necked region should correspond to the attainment of the ultimate elongation of the material within the neck itself. Plasticity results from slip processes taking place on planes of maximum shear stress. Consequently, changing the orientation of these planes results in different patterns of deformation. In the case of plane stress, maximum shear stress occurs on planes oriented approximately at 45° to the sample surfaces, Fig. 9a; it is apparent that the height of the plastic zone is proportional to the sample thickness, see the different plateau values for PVC in Fig. 7. On the

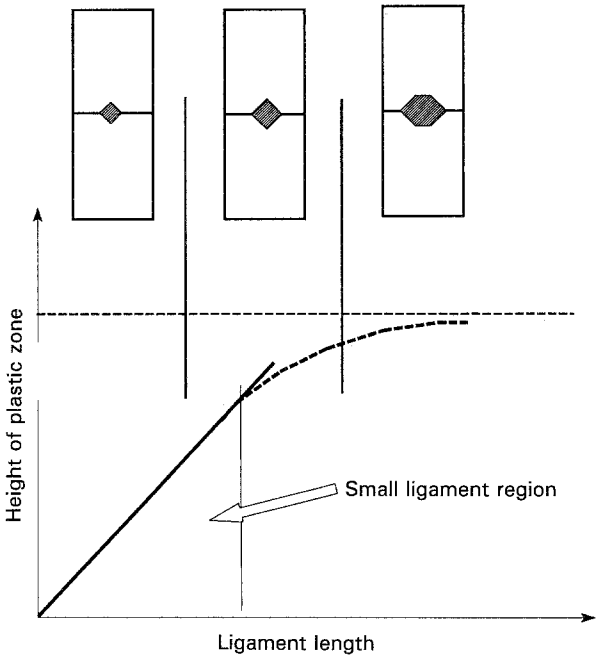

Figure 8 Schematic illustration of the height of plastic zone for different ligament length.

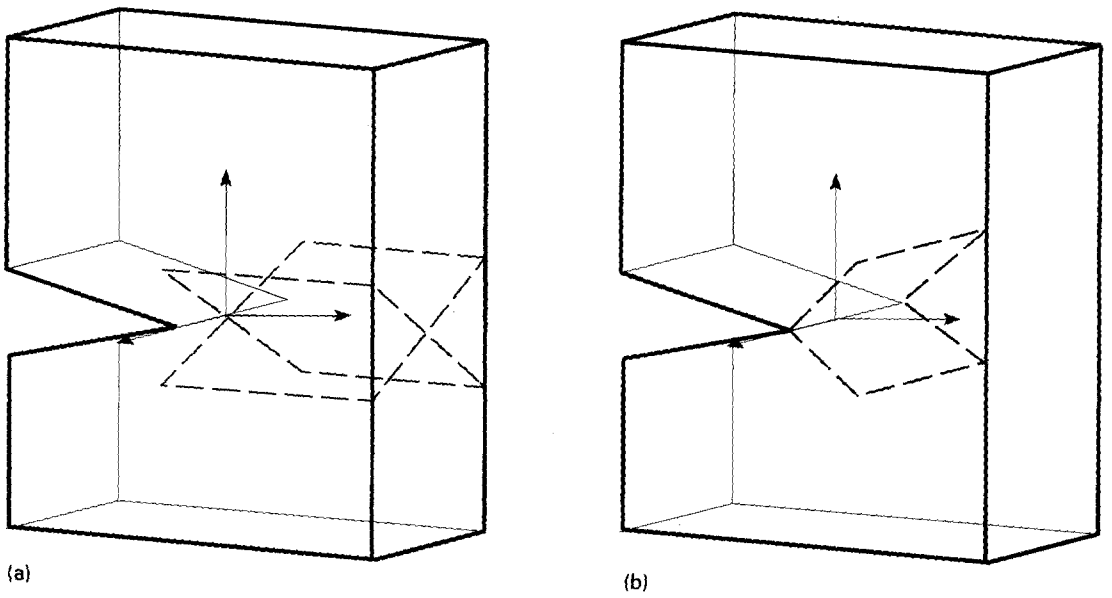

Figure 9 Orientation of maximum shear stress planes for (a) plane stress, (b) plane strain.

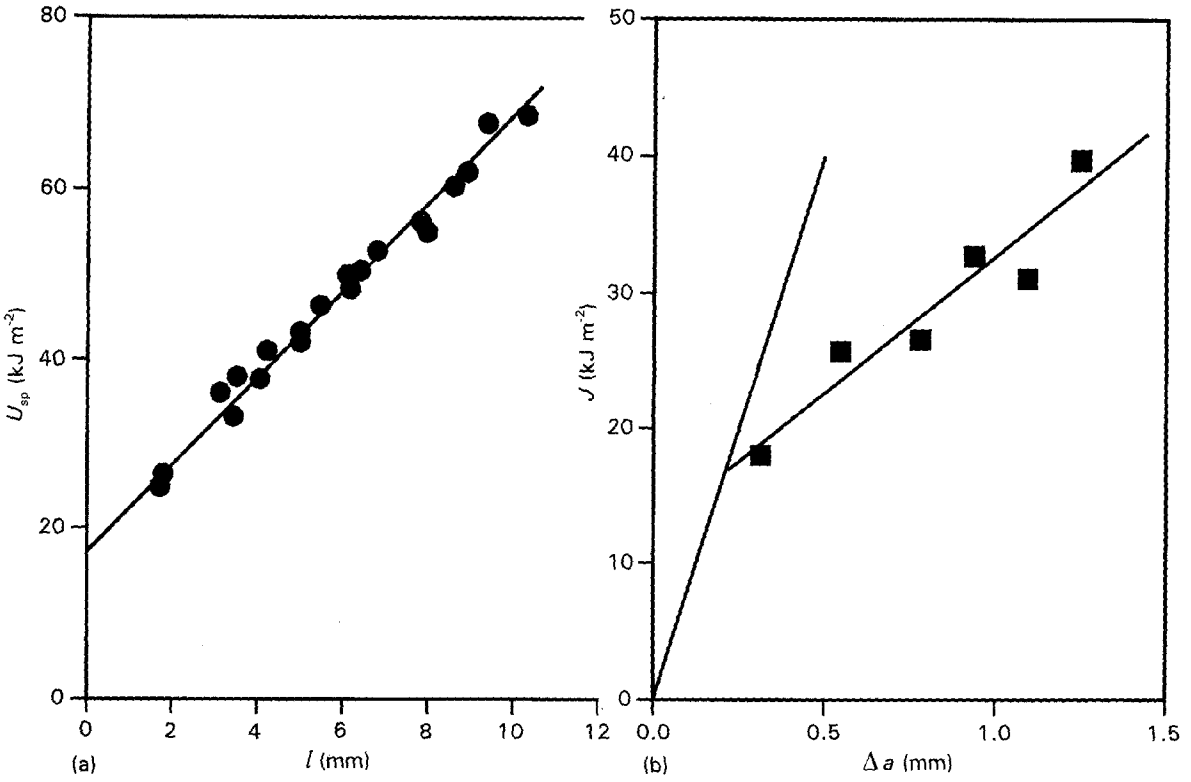

Figure 10 (a) Specific work of fracture versus ligament length and (b) work of propagation versus crack advancement for Ultranyl.

other hand, in plane strain the shear stress planes are normal to the sample surfaces, Fig. 9b, so that the height of the plastic region is independent of the thickness of the sample, as confirmed by the fact that no difference was found between the height of the plastic zone for 6 and 3 mm thick PVC samples in the small ligament region, Fig. 7.

Equation 10, referred to unit surface, becomes

$$u_S = \phi^* h^* + \bar{\phi}\alpha h \tag{12}$$

and by recalling that, at small ligaments, h is proportional to l (i.e. $h = \alpha' l$), Equation 12 can be recast as

$$u_S = \phi^* h^* + \bar{\phi}\alpha\alpha' l \tag{13}$$

This equation, which coincides with Equation 8 if $\phi^* h^* = u_E$, $\bar{\phi} = u_{NE}$ and $\alpha\alpha' = \beta$, predicts a linear dependence of the specific work on l; experimental data to support this conclusion, Figs 10a, 11a and 12a. An important difference between Equations 8 and 13 is, however, worth noting. In the original enunciation of the EWF model, the linear dependence was indicated to hold in the large ligament region, i.e. for ligaments exceeding the sample thickness by a factor of 3–5 in order to build up plane stress conditions in the ligament. Our reasoning indicates that the linear dependence should rather be sought at small ligaments (where h linearly depends on l) and that the

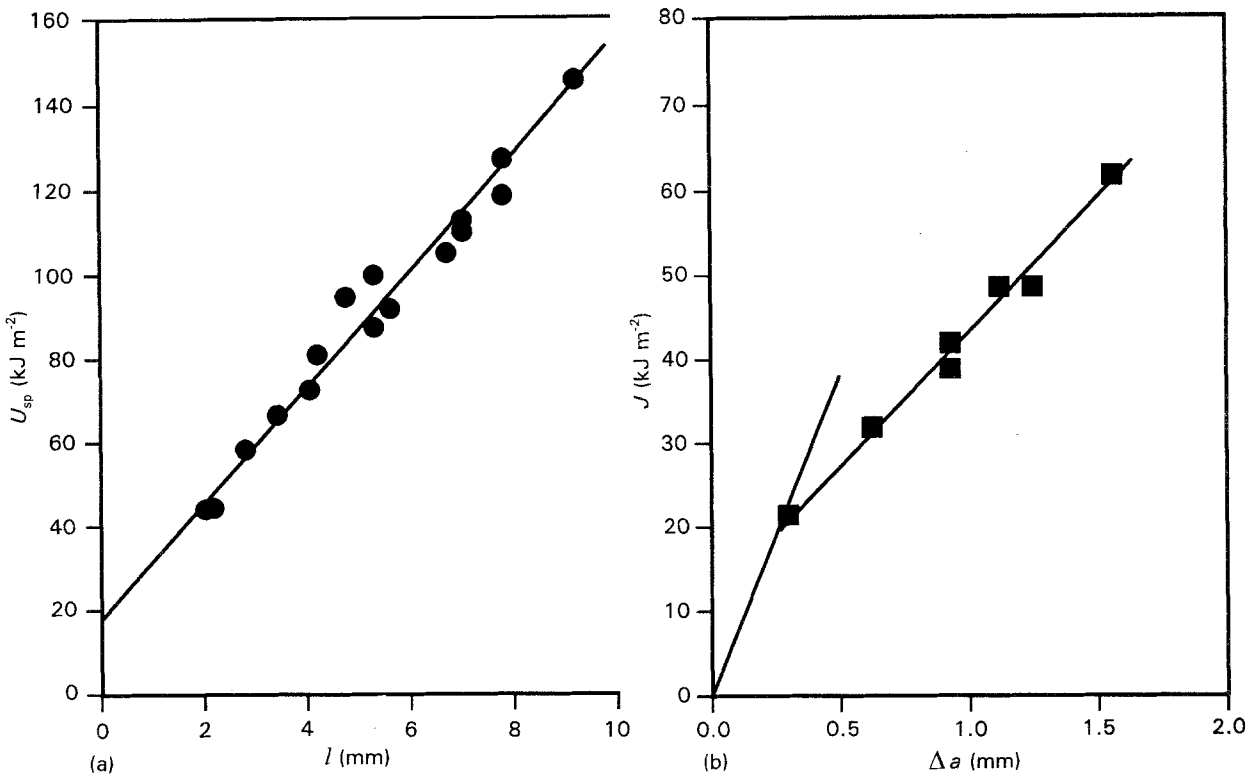

Figure 11 (a) Specific work of fracture versus ligament length and (b) work of propagation versus crack advancement for Orgalloy.

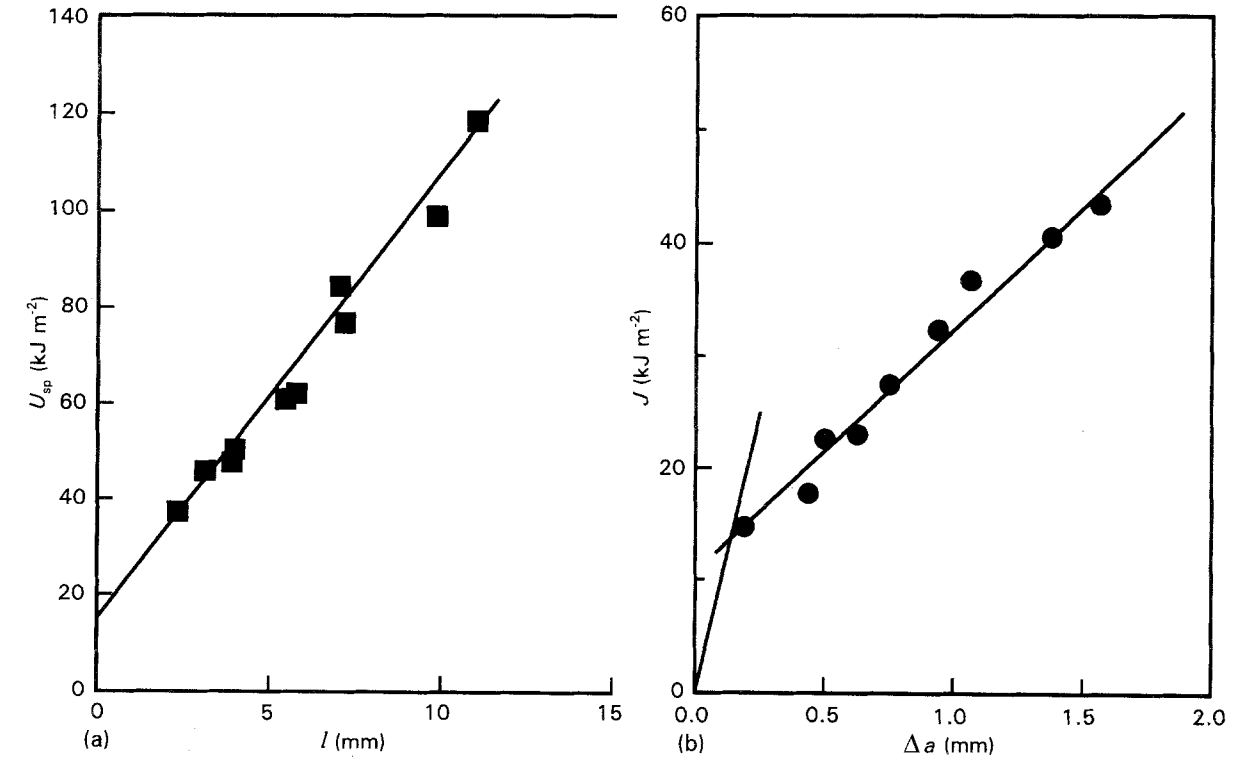

Figure 12 (a) Specific work of fracture versus ligament length and (b) work of propagation versus crack advancement for PVC.

transition from small to large ligament regimes is, in reality, dictated by the dependence of h on l which, in turn, is expected to reflect, at least in part, the post-yielding behaviour of the material (i.e. strain softening or strain hardening). The tendency of h to level off has the result that the rate of increase of u_S diminishes on enlarging l and the intercept on the ordinate axis increases. Consequently, no specific meaning can be given to the quantities u_E and u_{NE} as obtained by extrapolation from the large ligament region in u_S versus l plots. Much more interesting are the same quantities measured in the small ligament region, because they promptly reflect the processes taking place at the crack tip. From now on, the small ligament essential work of fracture will be indicated as u_{EC}. The overall dependence of the specific work of

fracture, u_S, on l is well represented, at least for ligaments not exceeding one-third of the sample width, by a power law of the type

$$u_S = Al^n \quad (14)$$

This was first observed by Salemi and Nairn [19] for toughened nylons and it seems to be a very general behaviour, as displayed in Fig. 13. These authors proposed using the power-law dependence as a tool to obtain data at large ligaments from measurements on small ligaments. In our view, the use of the power law should rather be reversed: improve the accuracy at small ligaments (it is, for instance, easy to deform in torsion samples with small ligaments while they are gripped in the test machine) utilizing a larger interval of the independent variable, l. The small ligament essential work of fracture, or the equivalent quantity defined here $\phi^* h^*$, has been identified [19] with the critical J-integral (at least for the specific thickness being tested). For all materials, we measured J as a function of crack advancement according to ASTM E 813-81, Figs 10b, 11b and 12b. Indeed, the coincidence of $\phi^* h^*$ with J_{IC} is rather convincing, Table I. Substantially, J_{IC} measures the work done in creating, at the crack tip, a plastic zone of thickness equal to the CTOD and of unit cross-section; consequently, $J_{IC} = \lambda\sigma_y$CTOD, where λ is an empirical parameter customarily set to 1. The essential work of fracture has the same meaning, i.e. the work done to develop the inner plastic zone of thickness h^* and of unit cross-section. $u_{EC} = \phi^* h^*$. The operative difference between J_{IC} and u_{EC} is that the former is obtained by the intercept between two lines that represent the fictitious and real propagation, whereas the latter is obtained by extrapolation to $l = 0$ the linear best fit to u_S/l data. There is, of course, no physical meaning in the work done to yield a ligament of vanishing length; it is only the approximation of Equation 9 made by Equation 13 to suggest this graphic procedure. The linear fit can, in fact, be meaningfully carried out only as long as $h > h^*$; therefore, it would be helpful to be able to extract h^* from h. According to Cotterell and Reddell [11] a plot of the displacement, ΔH, of a couple of fiducial marks, located outside the ligament area, Fig. 2, as a function of l has, for $l = 0$, a finite intercept identified as the opening displacement at the onset of crack propagation for the specific thickness being tested. We argue that the same procedure applied to ΔH taken at yield (in practice at the maximum of the experimental load/displacement curves), Fig. 14, rather than at failure, would give h^* with a better approximation. The difference between J_{IC} and u_{EC} is that in the first approach the transition from fictive to real propagation identifies the critical condition, whereas in the second approach the critical condition corresponds to yielding a ligament whose associated plastic zone is just h^* in thickness and $l^* = h^*/\alpha'$ in length. The work done per unit surface, is $u_{EC} \equiv \lambda\sigma_y h^*$ where λ measures the increase of plastic flow stress originated by the plastic constraint. From the plot of the flow stress versus l the value of the constraint factor, relative to l^*, can be estimated. In the case of Ultranyl we have

u_{EC} (as the intercept in the u_S versus l plot) $= 16\ \text{kJ m}^{-2}$

h^* (as the intercept in the ΔH_{YIELD} versus l plot) $= 0.28$ mm

λ (from the σ_{flow} versus l plot at $l = l^*$) $= 1.3$

σ_y (from uniaxial tensile tests) $= 45$ MPa

α' (from the h versus l plot) $= 0.48$

so that $\lambda\sigma_y h^* = 16.2\ \text{kJ m}^{-2}$ which coincides with the value of u_{EC}. The quantities u_{EC}, J_{IC} and $\lambda\sigma_y h^*$ can thus be considered equivalent to one another; this point is now under further evaluation for a number of different polymers.

It is of interest to note that, according to ASTM E 813-87, J is related to the crack advancement, Δa, according to a power law: $J = C_1 \Delta a^{C_2}$ and to support

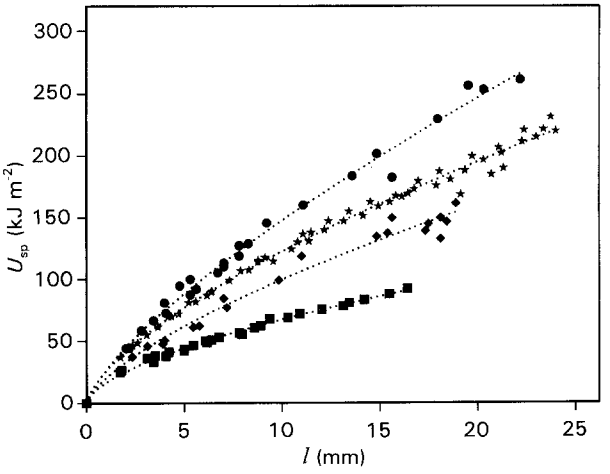

Figure 13 Power law best fits to specific work/l data for various polymers: (■) Ultranyl, (★) RTN66 [19], (◆) PVC, (●) Orgalloy.

TABLE I Comparison of critical parameters from work of fracture and J methods

Materials	u_{EC} (kJ m^{-2})	J_{IC} (kJ m^{-2})
Ultranyl	16	18
Orgalloy	20	18
PVC 6 mm	16	14
RTN66	21 [19]	20 [23]

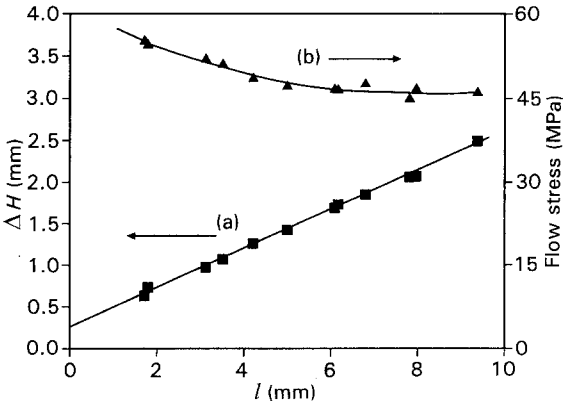

Figure 14 (a) Displacement at yield and (b) flow stress as function of ligament length for Ultranyl.

TABLE II Values of the exponent in the power laws for u_s and J methods

Materials	From u_s	From J
Ultranyl	0.57	0.52
Orgalloy	0.74	0.68
PVC 6 mm	0.69	0.68
RTN66	0.67 [19]	0.70 [23]

further the similarity between J and specific work of fracture, it can be pointed out that the two exponents of the power law, C_2 and n equation 14, are very close for the same material, Table II.

4. Conclusions

On increasing the remote load on DENT samples, the ligament yields due to the stress intensification caused by the local decrease in cross-section. Depending on the post-yield behaviour of the material, plasticity can localize or spread out. In the case of polymers, which strain-soften substantially, plasticity tends to remain confined in the ligament region. On further stretching the samples, fracture starts propagating and the work to fracture samples of semiductile materials is that required to produce the plastically deformed region that surrounds the fracture path. The height of the plastic zone increases with the ligament size; linearly at the beginning and with a tendency to level off at large ligaments. By assuming that the density of plastic work is made up of two terms, one evenly distributed within the plastic zone and the other localized in the vicinity of the fracture surfaces, it has been shown that a linear relationship exists between specific work of fracture and ligament length, provided the height of the plastic zone is in the linear regime. Extrapolation of specific work of fracture to $l = 0$, from the small ligament interval, gives values that coincide with the J_{IC}.

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Received 17 May 1993
and accepted 10 January 1994