Fracture energy analysis of single-edge-cracked isotropic ductile polyolefins

Min-Diaw Wang, E. Nakanishi*, Y. Hashizume and S. Hibi
Department of Materials Science and Engineering, Nagoya Institute of Technology, Nagoya 466, Japan

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Evaluation of energies for crack initiation and propagation is performed on isotropic low density polyethylene (LDPE) and polypropylene (PP). Loading–unloading loops were used to calculate the energies involved during cracking. For PP, an uncracked sample shows necking upon stretching and exhibits high toughness. It was found that, when loaded, the cracked PP sample also showed necking from the crack tip. As a consequence, a crack propagated inside the fully plastic deformed domain. While uncracked LDPE undergoes uniform deformation with strain hardening upon loading, no necking phenomenon was observed at the crack tip of the cracked sample. As the crack propagated, it had to go through the ligament. We suggest that the ligament area is an important fracture parameter in evaluating the fracture energy. In both cases, energies were divided by the ligament area before being related to the crack length increment. Thus, the potential necessary to create unit crack propagation is represented as energy per unit ligament area. As our results show, the fracture resistance of PP is higher than that of LDPE and this is related to the difference in toughness of these two materials.

(Keywords: fracture; plastic deformation; R curve)

INTRODUCTION

As polymeric materials become more and more promising in replacing metals for engineering applications owing to their high performance properties, study of the strength of polymers has received wide attention. For brittle polymers such as polymethylmethacrylate and polystyrene, application of linear elastic fracture mechanics (LEFM) has been proved to be successful[1]. One of the difficult tasks in studying the fracture of ductile polymers is to deal with materials showing large plastic deformation when the samples are loaded. This refers to the inelastic toughness fracture mechanics. Several methods have been proposed such as critical crack-opening displacement (COD), the path-independent J integrals[2], general fracture mechanics (GFM)[3] and the essential work theory[4]. For polymers above their glass transition temperatures, their mechanical behaviour becomes non-linear, inelastic and time dependent. In this case, the COD method seems to lose its merit as a valid criterion for polymer fracture. The J integral, GFM and the essential work theory, on the other hand, seem to deserve more interest.

J *integral*

Proposed by Rice[2] and further improved from theory into a practical experimental procedure by Begley and Landes[5], the J integral became one of the most widely accepted theories for non-linear fracture mechanics. As the theory originally stated, for an elastic material, not necessarily linear, containing a crack and under load, the J integral is defined by:

$$J = \int_{\Gamma} \left(W \mathrm{d}y - T \frac{\partial u}{\partial x} \mathrm{d}s \right) \tag{1}$$

where Γ is a curve surrounding the crack tip, W is the strain–energy density and T is the stress acting on the curve Γ. Rice has shown that, for the two-dimensional case, the J integral is independent of the path Γ. As shown in *Figure 1*, the J integral can be evaluated from a simple tensile experiment[6]. Clark *et al.*[7] proposed that it is possible to use one specimen to obtain the J integral *versus* crack length increment ($J - \Delta a$) curve by the compliance method. The J_{IC} fracture criterion in *Figure 1* refers to the onset of stable crack growth[6] while the R curve refers to stable crack propagation. This experimental procedure, originally proposed for metals, has been applied to polymers extensively by Williams[8].

Generalized fracture mechanics

Andrews[3] proposed the GFM that gave fracture criteria for solids with no limitations such as elastic, linear mechanical properties or small strain. According to GFM, the surface work F is defined as:

$$F = K_1(\varepsilon_o) a W_{o,\mathrm{crit}} \tag{2}$$

where $W_{o,\mathrm{crit}}$ is the energy input density at crack initiation, a is the initial crack length and $K_1(\varepsilon_o)$ is an empirical quantity. In the case of elastic materials, it is shown that the surface energy equals half of J_{IC}[9]. Details of the experimental procedure will be given later.

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* To whom correspondence should be addressed

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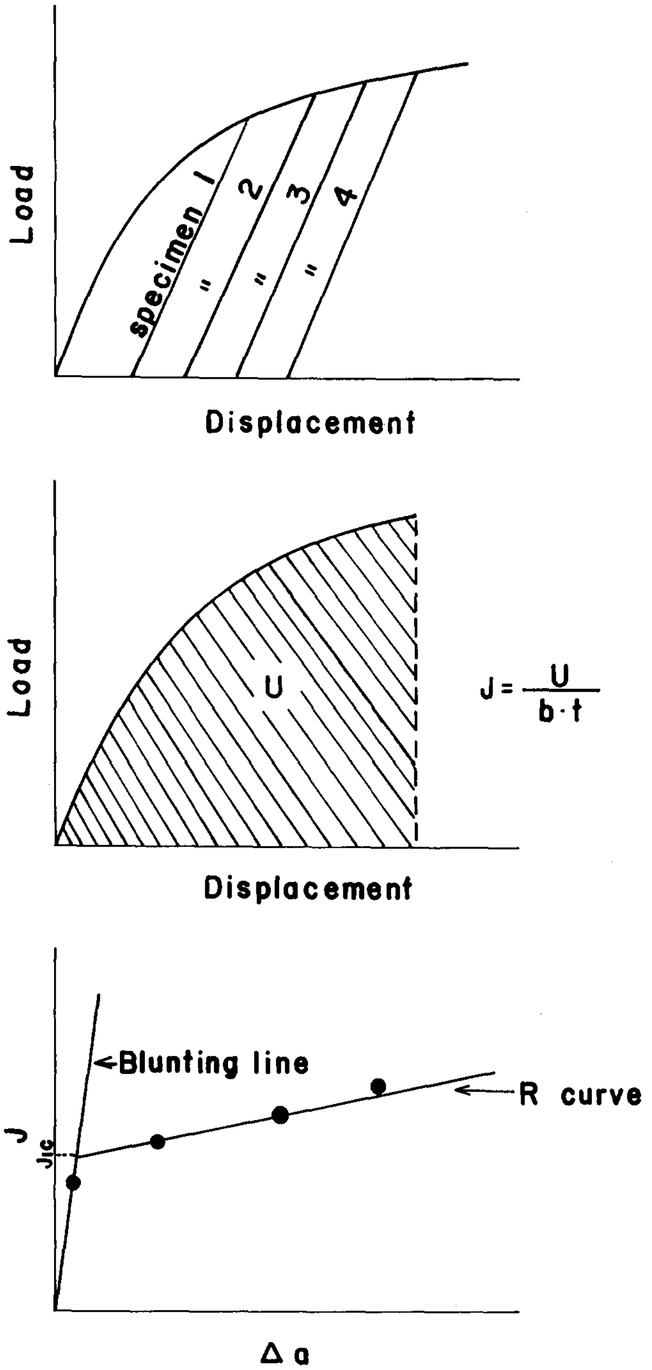

Figure 1 Evaluation of the *J* integral by the multiple specimen method[6]. *b* is the ligament length and *t* is the thickness of the specimen

Compared with the *J* integral, GFM is more flexible for materials exhibiting large deformation, while for stable crack propagation, it is not well defined.

Essential work theory

Proposed by Cotterell and Reddel[4], the essential work theory was originally designated for plane stress ductile fracture. For ductile materials with appropriate ligament length (b), the ligament will undergo full necking before crack initiation. For double deep-edge notched specimens with different ligament lengths, the load–displacement curve is as shown in *Figure 2b*. The total area under the load–displacement curve is defined as W_f, the total work to fracture the specimen. Cotterell and Reddel[4] proposed that W_f can be partitioned into the work, W_e, for crack growth inside the 'end zone' defined by Broberg[10] and the work, W_P, for plastic deformation that is not a necessary condition for crack growth. This W_P should be constrained only inside the 'outer plastic region', as shown in *Figure 2a*. W_P can be represented as $w_P(b^2t)$ where t is the specimen thickness, while W_e can be written as $w_e(bt)$. Then, a plot of $w_f(=W_f/bt)$ *versus* b as shown in *Figure 2c* will yield an intercept at w_e and the slope will be w_P. Mai and Cotterell[11] once claimed that this should be further divided into the fracture works for initiation ($w_{\rm I}$) and for propagation (w'_e). In their recent papers[12,13], they related w_e to $J_{\rm IC}$ again and defined w_e as the crack initiation energy.

As redefined by Landes and Begley[6], $J_{\rm IC}$ does not represent the initiation of crack growth but represents the start of stable crack growth, i.e., the onset of the R curve. $J_{\rm IC}$ might be close to the crack initiation energy, but the physical meaning should be distinguished. Also, the authors suggest that, for crack initiation in elastic–plastic fracture, it takes energy to initiate the 'end zone'. After that, for small crack propagation, it

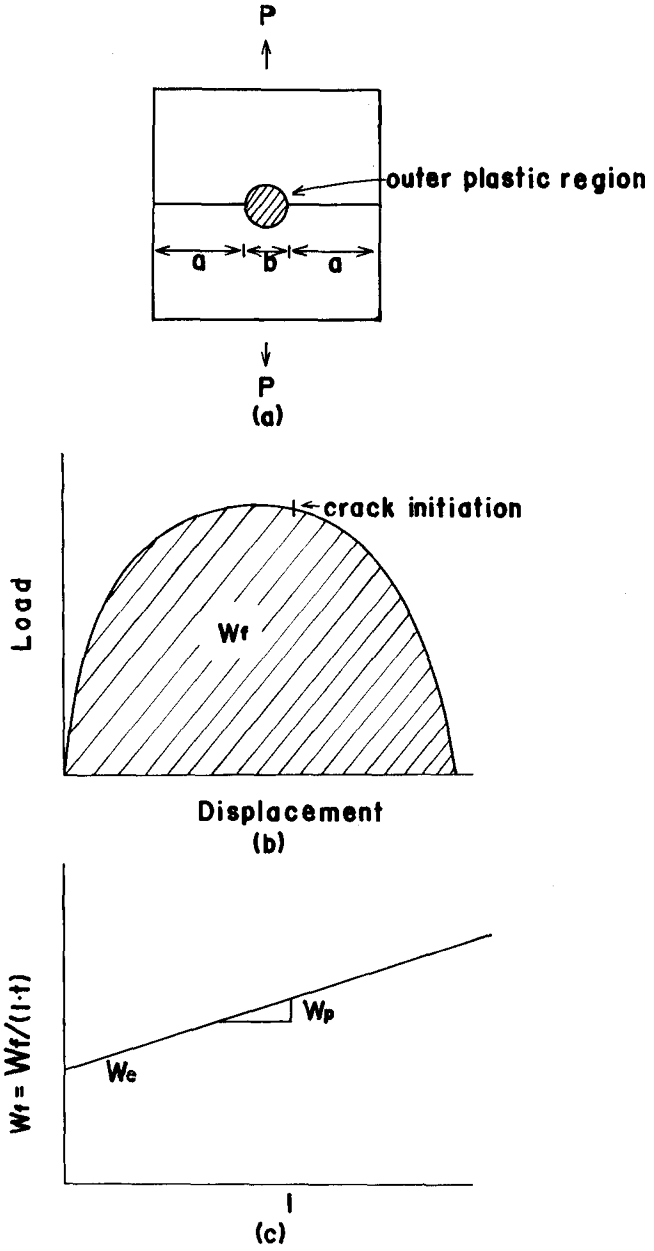

Figure 2 Experimental procedures in evaluation of the essential work theory[11]

undergoes elastic processes with small scale yielding and, during this period, the crack tip blunts. For further crack growth, it changes into an inelastic process and falls into the region of the R curve. The conclusion by Mai and Cotterell[11] seems to be more realistic to the authors. The essential work theory seems to be closer to the slope of the R curve intuitively.

Derived by Rice *et al.*[14] as shown in *Figure 1*, the J integral can be expressed as the energy from the load–displacement curve divided by the ligament area; even the original theory did not relate the J integral directly to the ligament. As the resistance to crack propagation comes from the ligament, it is reasonable to use the ligament area as a parameter in evaluating fracture energy without relating to the J integral theory. This has to be under the condition that the thickness of the ligament is known during the experiment. When GFM is used to obtain the surface energy, there is no special requirement about sample thickness.

As mentioned above, polymers above their T_g show non-linear behaviour, and large-scale yielding at the crack tips is a normal phenomenon. Using very thick samples to meet the requirement of plane strain is one method to make polymers suitable for applying the J integral theory. In this case, the effect from plastic deformation is not well understood. Also, as polymers show inelastic recovery behaviour, it is doubtful whether the methods used for materials that exhibit incremental plastic deformation can be applied to polymers. In this paper, we study the fracture energies of two ductile polymers that show plastic deformation with or without visible necking at room temperature. Instead of referring to complicated theory, data is analysed using simple physical intuition. Comparison of the results with the J integral and GFM is made and the energy consumed for plastic deformation is discussed.

EXPERIMENTAL

Two materials are used in this experiment. Low density polyethylene (LDPE) is supplied by Mitsubishi Petro. Chem. Co. with the trade reference PK-30. The yield stress is 5.67 MPa and the maximum stress is 19.3 MPa. This corresponds to a flow stress (σ_{fs}) of 12.5 MPa. Polypropylene (PP) is supplied by the Mitsui Toatsu Chem. Co. with the trade reference JH-G. Both were hot pressed at 200°C under pressures of 490 MPa and 980 MPa, respectively. Water quenching was used and the samples were cut to the dimensions shown in *Figure 3*. The L/W ratio was kept to 3 to avoid the grip effect[15]. The stretching speed was 0.4 mm min^{-1} to avoid thermal and dynamic effects. The loading–unloading process was carried out after a certain propagation of the crack. As shown in *Figure 4*, for point A and point B, no recoverable energy is imparted into the sample. That is to say, the mechanical energies of A and B are the same. The increment of the crack length from A to B is Δa. It is necessary for A to go through the path AA'B'B to reach to B, but, if unloading takes place at A', the unloading curve is A'CA. In consequence, it takes the energy represented as the shaded area to cause Δa crack growth.

PP

For PP, necking at the crack tip was observed immediately after the samples were loaded and it expanded quickly as the crack propagated. As shown in *Figure 5*, necking kept expanding from the crack tip towards the other end of the ligament. After necking passed through the ligament, it continued to expand in the stretch direction and became larger as the crack propagated. The thickness of the necking area was found to be quite uniform and constant. This can be explained by considering that the necked area was extended to the natural draw ratio[16] and the stress of the necked domain was almost constant. The energy was then considered to be used to drive the crack and to expand further necking. In order to evaluate the crack propagation energy with

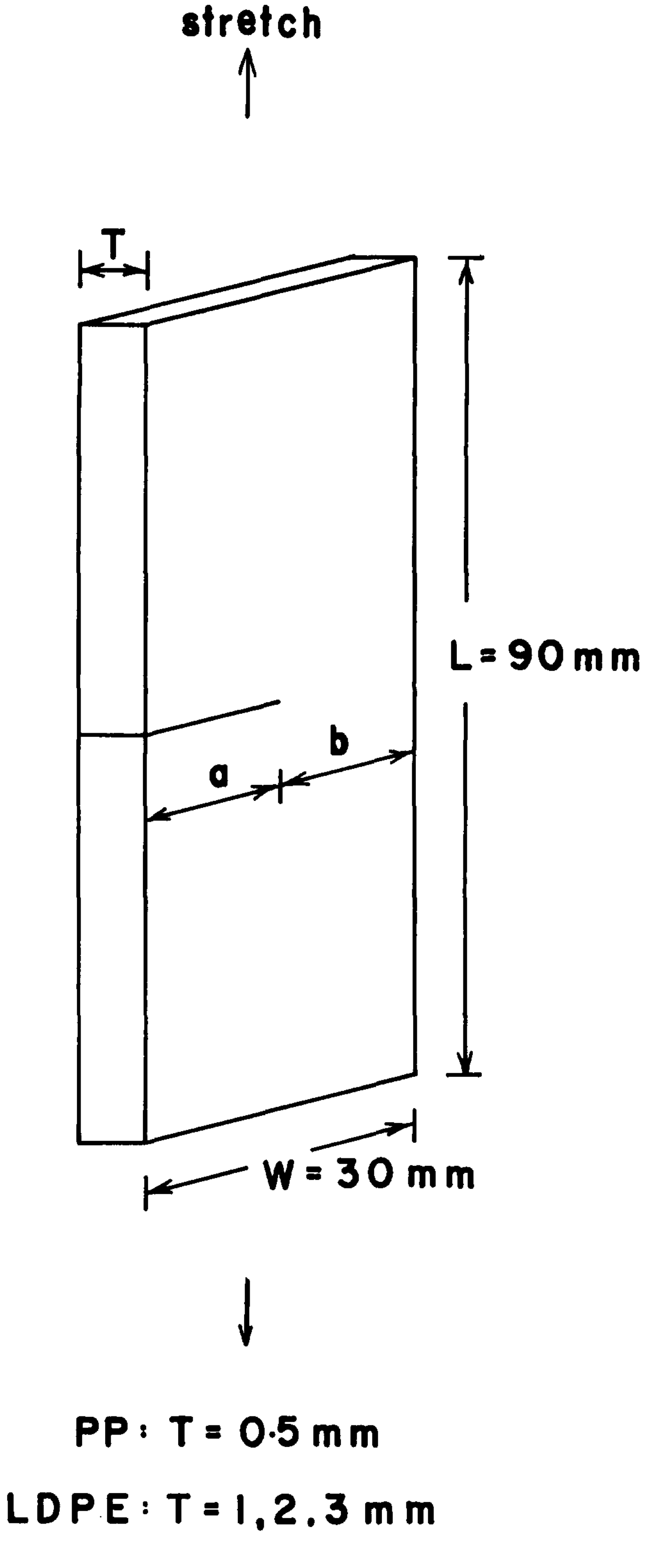

Figure 3 Schematic representation of specimen dimensions

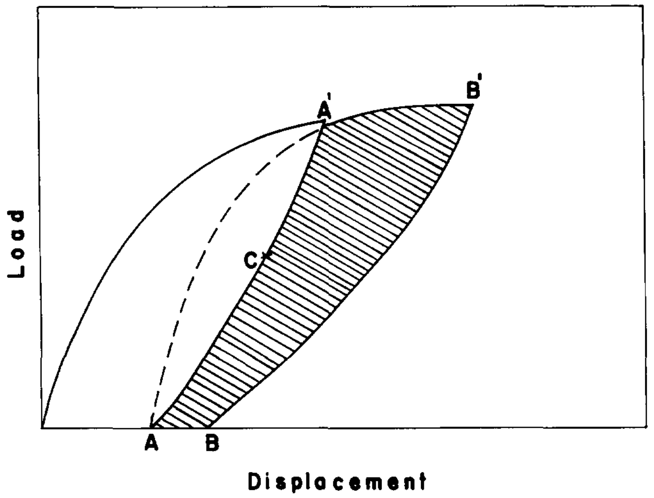

Figure 4 Schematic representation of the loading–unloading process. A and B are at the unloaded state

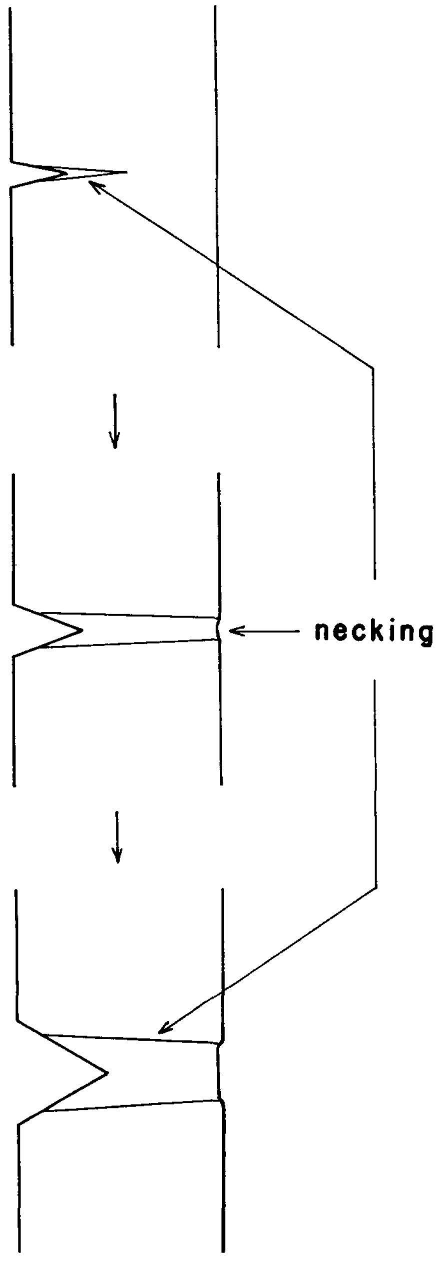

Figure 5 Expansion of necking with crack propagation of PP. The crack propagates before necking goes through the ligament

constant ligament thickness, we focused on what happened after the necking went through the ligament. The area of each loading–unloading cycle was calculated. Propagation of the crack and expansion of the necked area were monitored by a camera. Initial crack lengths were from 3 to 21 mm at 3 mm intervals. This experimental procedure was designed before the authors referenced the work of Cotterell and Mai[11–13].

PE

While LDPE did not show necking behaviour, the increment of the plastically deformed area during crack propagation could not be evaluated. In this case, samples with different initial crack lengths were loaded and then unloaded after every 1 mm stretching until the crack length increment reached 2 mm. Propagation of the crack was monitored by a camera. Samples with thicknesses of 1 mm, 2 mm and 3 mm were used to check the effect of thickness. The energy for crack propagation was calculated as described above. After the sample was unloaded, the thickness of the whole ligament did not show significant change. The initial crack lengths ranged from 6 mm to 21 mm, which corresponded to a/w, the crack length to the width of the specimen, with ratios ranging from 0.2 to 0.7.

In order to obtain the surface energy (the energy for crack initiation), the experimental procedure of GFM was applied to both materials. Initial crack lengths ranging from 3 mm to 21 mm with 3 mm intervals were loaded until initiation of crack growth was observed. The load–displacement curves were recorded.

RESULTS AND DISCUSSION

It is known that the energy to initiate a crack is not independent of the initial crack length; neither is the energy for crack propagation. In LEFM, the stress distribution σ_{ij} around the crack tip can be expressed as:

$$\sigma_{ij} = \frac{K_{\mathrm{I}}}{\sqrt{2\pi r}} \cos\frac{\theta}{2} f(\theta) \qquad (3)$$

for mode I deformation. Here K_{I} is the stress intensity function and is a function of the crack length and the applied stress. For longer cracks, K_{I} is larger so that the stress at the crack tip is higher. When the stress reaches some level, for example, the ultimate stress, the crack begins to move forward. Equation (3) seems to be independent of dimension. In practice, the effect of the configuration of the sample is involved in the applied stress and in turn affects K_{I}. In consequence, σ_{ij} is not really independent of the configuration. As the crack begins to extend or undergoes stable propagation, it has to go through the ligament. If no other dissipation of energy occurs very far from the crack and off the crack plane, the resistance of crack growth is strongly related to the ligament area. As a consequence, the ligament area should be considered as an important factor in the energy analysis.

Fracture resistance

PP. It can be seen in *Figure 5* that plastic deformation due to necking in front of the crack tip is large and the energy consumed by plastic deformation cannot be ignored. If the total energy during every unit crack propagation is the area of one loading–unloading cycle and is defined as ΔU_t, then ΔU_t is considered to consist of two parts. One is the energy for crack propagation in the necked area, ΔU_c, and the other is the energy for necking propagation, ΔU_p. If the increment of necking area during unit crack propagation is ΔA and the energy for unit volume of PP to undergo plastic deformation is ω_p, ΔU_p can be expressed as the following:

$$\Delta U_p = \omega_p \Delta V = \omega_p \Delta A t \qquad (4)$$

where t is the thickness of the necked area. Then ΔU_t can be expressed as:

$$\Delta U_t = \Delta U_c + \omega_p \Delta A t \qquad (5)$$

By dividing both sides of equation (5) by the ligament area (bt), equation (5) is rewritten as

$$\Delta U_t/(bt) = \Delta U_c/(bt) + \omega_p(\Delta A/b) \qquad (6)$$

In equation (6), $\Delta U_c/(bt)$ is the energy for unit crack propagation per unit ligament area under stable conditions and b is the remaining ligament length of the necked specimen. By plotting $\Delta U_t/(bt)$ *versus* $(\Delta A/b)$, the intercept should be $\Delta U_c/(bt)$, while the slope corresponds to ω_p. In order to have the thickness of the ligament equal to that of the necked area so that equation (6) can be established, data were analysed after necking passed through the ligament. This was achieved after the crack propagated about 2 to 3 mm. Shown in *Figure 6* are the experimental results. The straight line is obtained by least squares linear regression of the data. The intercept, $\Delta U_c/(bt)$, is found to 88.6 kJ m^{-2} for crack propagation and the slope, ω_p, is 73 MJ m^{-3}. Scattering of the data might be due to thickness non-uniformity and to a stress hardening effect inside the necked area. As all crack propagation is about 1 mm, $\Delta U_c/(bt)$ can be considered to be out of the range of crack initiation. Instead of dramatic failure, stable and slow crack growth was observed. $\Delta U_c/(bt)$ was thus interpreted as the energy per unit plastically deformed ligament area to process unit crack propagation under stable conditions.

Saleemi and Nairn[17] used double edge-cracked PE to evaluate $\Delta U_c/(bt)$ by the essential work theory. They also suggested that the $\Delta U_c/(bt)$ value corresponded to J_{IC} in the J integral theory. As our experiment was not limited to initial crack propagation, we would prefer suggesting that the $\Delta U_c/(bt)$ value corresponds to the slope of the R curve with the physical meaning mentioned above. This value also represents the fracture resistance of PP. The authors would like to emphasize the difference in both experimental procedure and interpretation of results between this work and the essential work theory.

An increase of the thickness of samples might give some restriction upon plastic deformation, and this might lead to the fact that energy consumed due to plastic deformation cannot be evaluated. Also, in order to meet the true plane strain condition of this material, it requires more experimental data with very thick samples that are not practically feasible for laboratory use. Such a thickness is to be used to constrain the plastic deformation. The necessary thickness might be larger than that defined in ASTM E399.

LDPE. As LDPE shows different deformation behaviour, the method applied to PP cannot be used here. Large scale yielding at the crack tip front was confirmed in our previous work[18]. The crack increment Δa was checked from the photographs and energy was calculated as the area in each cycle. The energy was divided by the ligament area and then was plotted as a function of Δa. *Figure 7* shows the results for the LDPE samples. The straight line with the smaller slope is obtained by best fitting of the data. For samples with 1 mm thickness, the data fit this straight line very well at small crack growths. This straight line possesses the physical meaning as the constant energy for stable crack growth per unit ligament area. As Δa becomes larger than 0.7 mm, the data begin to deviate from a straight line; that is to say, the energy per unit ligament area for unit crack growth is no longer a constant. Our previous work[18] on LDPE shows that as the ratio of plastic zone to ligament length becomes larger than 24%, the plastic zone is no longer predictable by the model proposed by us. One of the reasons lies in the fact that the plastic deformation in front of the crack becomes large not only in the crack plane but also expands to areas far from the crack. This is similar to the case of PP. Plastic deformation far from the crack is not a necessary condition for crack propagation. The energy dissipated by this kind of plastic deformation is considered equal to the part above the straight line in *Figure 7*. For 2 mm

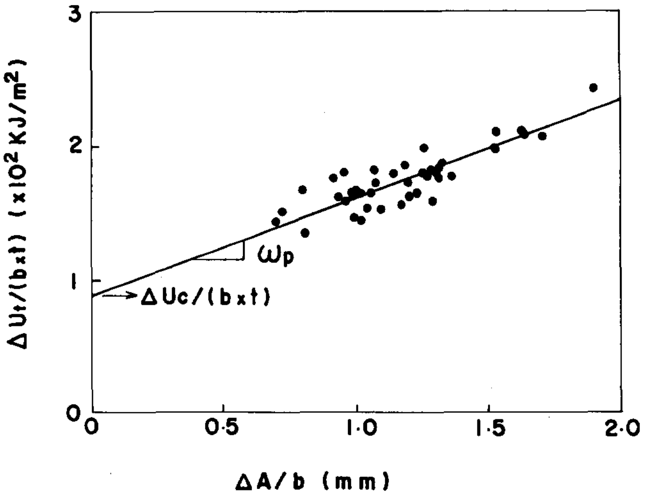

Figure 6 $\Delta U_t/(bt)$ *versus* $\Delta A/b$ of PP. The slope of the straight line is ω_P while the intercept is $\Delta U_t/(bt)$

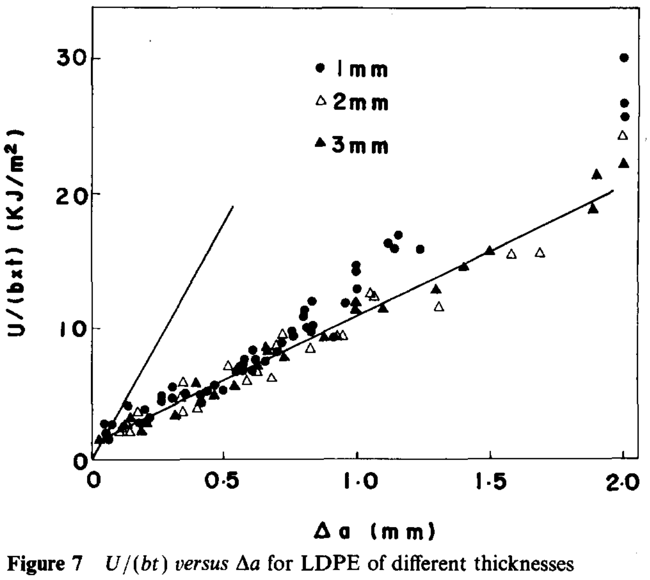

Figure 7 $U/(bt)$ *versus* Δa for LDPE of different thicknesses

and 3 mm samples, the same phenomenon was observed but at larger Δa. This might be due to the larger constraint from the thickness so that an effect from plastic deformation shows up after larger crack increments.

It is interesting to find that, when data began to deviate from the straight line, this became dependent on the crack length. An example is shown in *Figure 8* for 1 mm samples with large crack growth. It can be seen that samples with larger crack lengths deviate more from a straight line. This can be explained by assuming that a longer crack results in a higher stress concentration. This, in turn, leads to a larger plastically deformed area. The straight line in *Figure 7* can then be interpreted as the stable crack growth line and as the fracture resistance. The slope of this line is 10 kJ m^{-2}. Compared with PP, LDPE possesses smaller fracture resistance. As PP is a material with high toughness, it is not surprising to see that PP also shows higher resistance to fracture. It has to be kept in mind that, in both cases, the crack propagates in the plastically deformed domain.

For LDPE, the smallest energy evaluated is about 1.8 kJ m^{-2}, while the intercept of the straight line on the y-axis is about 1.4 kJ m^{-2}, which is close to the results by Barry and Delatycki[19] with a similar strain rate. The straight line connecting the origin and the least energy point in *Figure 7* shows a slope larger than $2\sigma_{fs}$, the ideal slope of the blunting line in *Figure 1*. This implies that the critical blunt angle for stable crack growth is more than 45°. This is a normal phenomenon for ductile polymers that usually show larger crack-opening displacement than predicted. In the case of PP, the blunting line could not be obtained because the data were taken after large crack growth.

Crack initiation

The resistance to stable crack propagation in a plastically deformed area has been discussed. Another important fracture parameter is the energy for crack initiation. It is shown in *Figure 7* that the intercept is about 1.4 kJ m^{-2}. At this point, the crack just began to grow. GFM[3] is utilized to confirm this and to evaluate the energy for PP crack initiation. Shown in *Figure 9* is an outline of the procedures for calculating the surface energy. Samples with different initial crack lengths were stretched until the start of crack growth and plotted on the same scale as in *Figure 9a*. Uncracked samples were used as the basis to obtain various W_0 (input energy density). Energies for samples to pass from zero crack length to various crack lengths at each W_0 were obtained as $\varepsilon_o - \varepsilon_i$. These values were plotted as a function of crack length and derivatives at different crack lengths were taken as shown in *Figure 9b*. The empirical quantity K_1 in equation (2) was defined as the slope in *Figure 9c*. The surface energy was then derived as in *Figure 9d.*

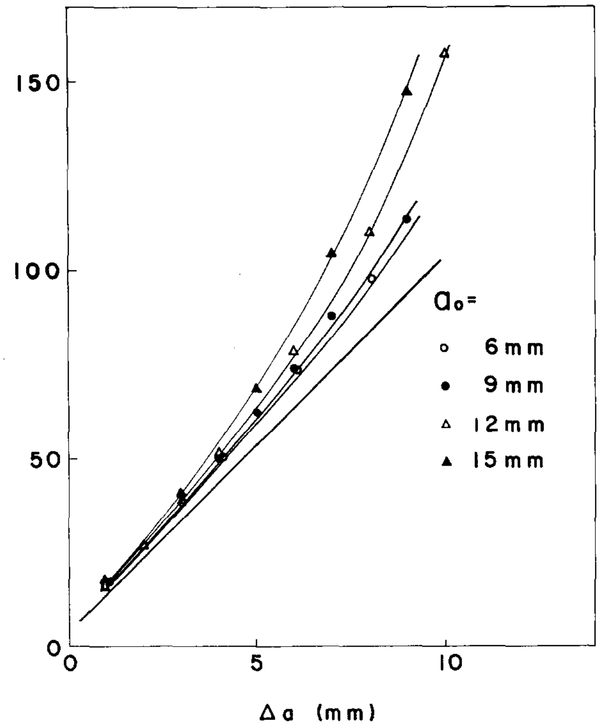

Figure 8 Effect of initial crack length on the fracture toughness of LDPE. The straight line corresponds to the stable crack growth line in *Figure 7*

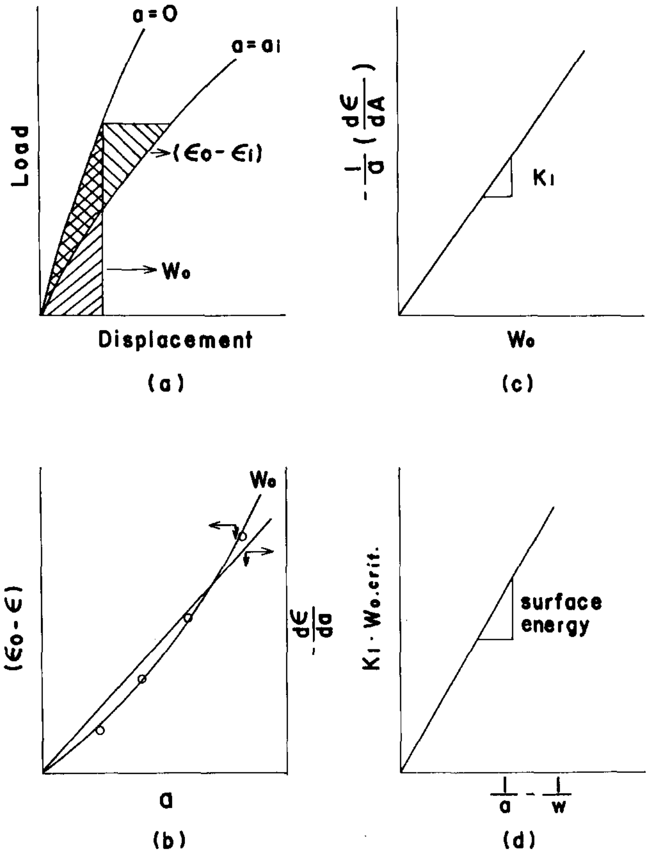

Figure 9 Experimental procedures in evaluating the surface energy[3]

The results for LDPE and PP are shown in *Figure 10* and *Figure 11*, respectively. The surface energy of LDPE is about 0.68 kJ m^{-2} while that of PP is about 3.24 kJ m^{-2}. For LDPE, half of the intercept in *Figure 7*, which possesses the same physical meaning as the surface energy, is very close to the slope in *Figure 10.* According to Andrews[3], these values refer to the energies for crack initiation. The surface energy and J_{IC}, even with different physical meanings, should be very close to each other in value. If the surface energy is taken as half of J_{IC}, as suggested by Andrews and Bhatty[9], the thickness requirement to meet the plane strain condition would be 2.7 mm for LDPE according to the following equation[6]:

$$t \geqslant 25(J_{IC}/\sigma_{fs}) \qquad (7)$$

Then the 3 mm samples in this experiment should be able to be interpreted with the J integral theory. The straight

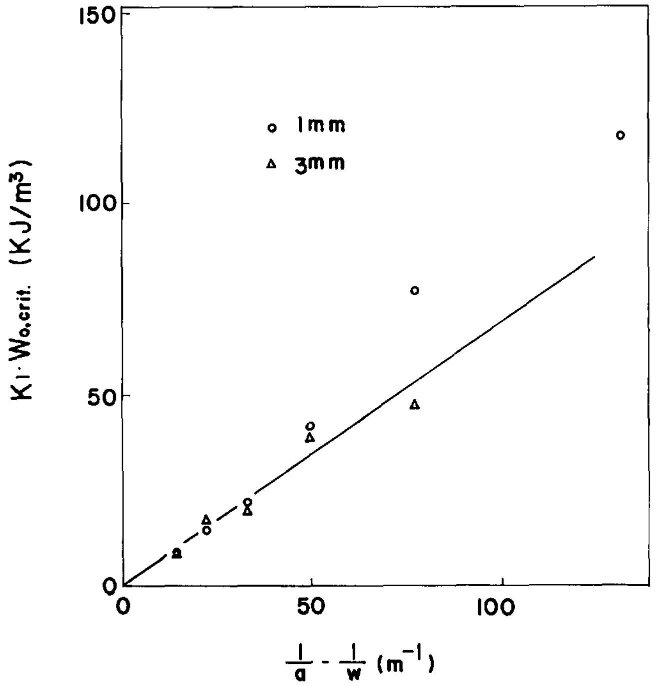

Figure 10 Plot of $K_1 W_{o,crit}$ *versus* $(1/a - 1/w)$ for LDPE for 1 mm and 3 mm samples. w is the width of the specimen

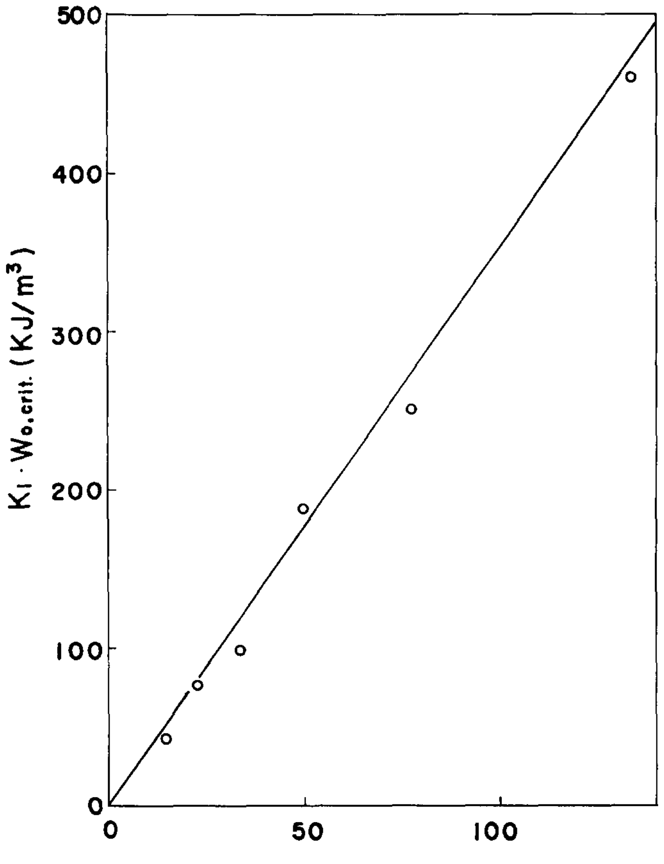

Figure 11 Plot of $K_1 W_{o,crit}$ *versus* $(1/a - 1/w)$ for PP. w is the width of the specimen

line in *Figure 7* is then equal to the R curve. As the stable crack growth line in *Figure 7* is the same for both 1 mm and 3 mm samples, this implies that, for 1 mm samples at small crack growth, it is still possible to characterize the R curve. The loading–unloading process is very clear in its physical meaning in that non-linear and inelastic effects were considered by unloading. Also, energy stored as elastic energy in the sample far from the crack was released. It is found that, for the same initial crack length, the unloading curves at different crack growths were very different in slope and incremental plastic deformation was not the case for these kinds of polymers. Application of the compliance method, which is good for metals with incremental plastic deformation behaviour, to polymeric materials should be made very carefully if not taking this into consideration.

CONCLUSION

Fracture energy analysis of two ductile polymers has been performed. A loading–unloading process was applied to obtain energies for crack propagation. In the case of polypropylene, procedures similar to the essential work theory were used to evaluate the energy (U_c/bt) for unit crack growth per unit plastically deformed ligament area. In this experiment, the necked area did not stay constant so that the essential work theory could not be applied. We consider $U_c/(bt)$ as the energy for stable crack propagation in a plastically deformed region. Consistent results showing a similar crack initiation energy and R curve were obtained for LDPE specimens with different thicknesses. GFM was utilized to reconfirm the crack initiation energy (twice the surface energy) of LDPE and PP. As fracture involves deformation and cleavage of a continuous domain, it should be strongly related to the toughness of materials. In this work, PP shows higher fracture resistance than LDPE and this relates to the higher toughness of PP than LDPE. The blunting line of LDPE was found to have a slope larger than $2\sigma_{fs}$. This indicates that the critical crack opening angle was larger than 45°, which is the ideal critical blunt angle at the crack tip.

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