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On the essential and non-essential work of fracture of biaxial-oriented filled PET film

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The plane-stress fracture toughness of a biaxial-oriented, filled poly(ethylene terephthalate) (BOPET) film was determined by the essential work of fracture concept using tensile-loaded deeply double-edge notched (DDEN-T) specimens. Results suggested that the essential work of fracture (w_e) is a material parameter, whereas the non-essential or plastic work (βw_p, where β is the shape factor of the plastic zone) depends on the testing conditions. Various approaches were used to assess w_p either explicitly (uniaxial tensile and trouser tests) or via estimation of β [by infra-red thermography (IT)]. IT seemed to be the most reliable technique for this purpose. IT also answered the question whether necking precedes crack growth or these processes occur simultaneously during specimen loading. The out-of-plane type deformation (mode III) in the trouser test resulted in markedly lower w_e and higher w_p values than those determined by in-plane (mode I) tests on DDEN-T specimens. The difference was attributed to the microstructure-related anisotropy of the PET film caused by biaxial drawing. Copyright © 1996 Elsevier Science Ltd.

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INTRODUCTION

Much effort is currently being made to assess the toughness of polymers using the concepts of both linear elastic fracture mechanics (LEFM) and elasto-plastic or post-yield fracture mechanics (PYFM)[1,2]. For ductile polymers and related blends, two approaches of PYFM seem to be most straightforward: the *J*-integral approach[1-4] and the essential work of fracture[1,3-5]. The latter concept differentiates between the essential work (W_e, required to fracture the polymer in its process zone) and non-essential or plastic work (W_p, consumed by various deformation mechanisms in the plastic zone), as indicated in *Figure 1*. The total work of fracture (W_f) is composed of the two above terms:

$$W_f = W_e + W_p \tag{1}$$

Taking into consideration that W_e is surface-related whereas W_p is volume-related, W_f can be given by the related specific work terms (i.e. w_e and w_p, respectively):

$$W_f = w_e l t + \beta w_p l^2 t \tag{2}$$

$$w_f = \frac{W_f}{lt} = w_e + \beta w_p l \tag{3}$$

where l is the ligament length, t is the thickness of the specimen and β is a shape factor related to the form of the plastic zone.

Based on equation (3), the specific essential work of fracture (w_e), being a material parameter, can be easily determined by reading the ordinate intercept of the linear plot of w_f *versus* l. The main advantage of the essential work approach is that various specimen geometries can be used for the evaluation of w_e and w_p. Several results hint that w_e is rather insensitive to changes in the microstructure of the polymer, while w_p reflects them more properly[6]. This statement can be explained by the fact that toughness-controlling energy absorption mechanisms (such as crazing, cavitation, shear yielding and their combination) are taking place in the plastic zone. The explicit determination of w_p, however, necessitates the knowledge of the shape factor β [see equations (2) and (3)].

The plastic work dissipated per unit volume (w_p) in the material can be defined in three ways:

1) by tensile tests to failure on dumb-bell specimens in which case w_p is defined as the ratio of the total failure energy to the volume of the fully yielded region[7];
2) by considering the shape of the plastic zone in deeply double-edge notched tensile (DDEN-T) specimens[1,7]; and
3) by determining w_p directly from the trouser tests (*Figure 1*)[3,8].

Assessment of w_p from tensile tests on dumb-bell and DDEN-T specimens is rather simple, provided that the yielded, necked region can be precisely defined.

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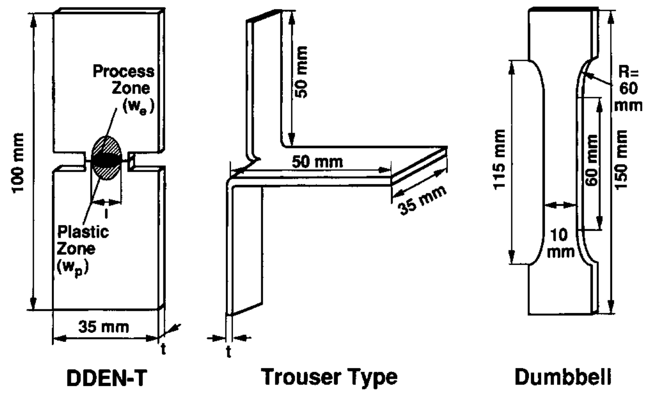

Figure 1 Designation and size of the specimens used

However, this is not generally the case with filled and/or oriented polymers where both necking and stress-whitening in the plastic zone can hardly be resolved. A further related question is whether or not w_p derived from the trouser tests (out-of-plane or mode III deformation, see *Figure 1*) matches that from in-plane (mode I) tensile tests.

The objective of this work was to determine the fracture toughness of biaxial-oriented, filled poly(ethylene terephthalate) (BOPET) film by applying the essential work of fracture concept. This material was selected based on the reasoning that a filled BOPET film may represent the 'worst example' for this purpose, owing to its restricted deformability (due to molecular orientation and filling) and opacity (hampering determination of the stress-whitening and thus estimation of the shape of the plastic zone).

EXPERIMENTAL

Biaxial-oriented, chalk-filled (filler content ~ 12 wt% according to thermogravimetric analysis) PET film of 250 μm thickness (Melinex®, ICI, Wilton, UK) was used in this study. DDEN-T specimens with a width of 35 mm and overall length of 100 mm (clamped length 70 mm; see *Figure 1*) were first punched and notched by scissors. The free ligament length was set in the range $l = 5$ to 20 mm. Tensile loading of the specimens, with their length axis along (L) or transverse (T) to the machine direction, was conducted at 1 and 20 mm min^{-1} on a Zwick 1445 universal testing machine (Ulm, Germany) at room temperature (r.t.). At least three specimens were investigated for every ligament. Data reduction [equation (3)] followed the recommendations of the ESIS TC-4 group[7].

For the determination of w_p, all the aforementioned methods—i.e. tensile testing on dumb-bells, *in situ* assessment of the plastic zone and direct evaluation from trouser tests—were used. Tensile loading of dumb-bell specimens (No. 3, according to DIN 53455 standard) was performed at r.t. in the cross head speed (v) from 1 to 100 mm min^{-1}.

The opacity of the PET film did not allow us to follow the development of the plastic zone by transmission light microscopy. This difficulty was circumvented by applying the technique of infra-red thermography (IT; Hughes thermal video system, Portland, OR, USA). IT frames were taken on videotape (continuously or at selected points of the loading curve) during loading of the DDEN-T specimens. The aim of applying this technique was to obtain a mapping of the relative temperature rise in the ligament region, so an arbitrarily chosen emission factor ($E = 0.9$) was set. IT pictures served for estimation of both the shape and extension of the plastic zone.

Tearing of the PET film was performed at r.t. with $v = 1$ and 20 mm min^{-1}, using trouser-type specimens (of the same overall dimensions as the DDEN-T specimens, i.e. 100×35 mm^2) with a leg of 50 mm (*Figure 1*).

In addition, the necked ligament area of the failed specimens was studied both by differential scanning calorimetry (d.s.c.; DSC-30, Mettler-Toledo, Greifensee, Switzerland) and Fourier transform infra-red spectroscopy (*FT* i.r. NicPlan®, Nicolet, Madison, WI, USA) in order to check whether or not PET underwent strain-induced crystallization during necking.

RESULTS AND DISCUSSION

Effects of specimen orientation and crosshead speed

Figure 2 depicts the w_f *versus* l curves for the DDEN-T specimens, tested in both the L- and T-directions, at crosshead speeds $v = 20$ and 1 mm min^{-1}. w_f was computed from the area under the force–elongation ($F - x$) curve registered during loading. F–x curves were similar to one another (*Figure 3*), demonstrating that the basic requirements of the essential work of fracture concept are met. The fact that practically the same regression line fits the results obtained on L- and T-specimens suggests that BOPET was drawn equally in both machine and transverse directions (balanced BOPET).

Reducing the crosshead speed to $v = 1$ mm min^{-1} had no effect on w_e but markedly decreased the slope of the regression line, i.e. βw_p [equation (3)]. This was due to an alteration in the mechanical response of the BOPET with v (associated with distortion in the related F–x curves, *Figure 3*). With unoriented PET films no change in the βw_p term was observed[5], therefore the above finding is probably related to the biaxial orientation of the PET film studied.

As determined from the ordinate intercept, w_e was in the range 45.0 to 47.0 kJ m^{-2} irrespective of v. This w_e range is below that reported by Chang and Williams

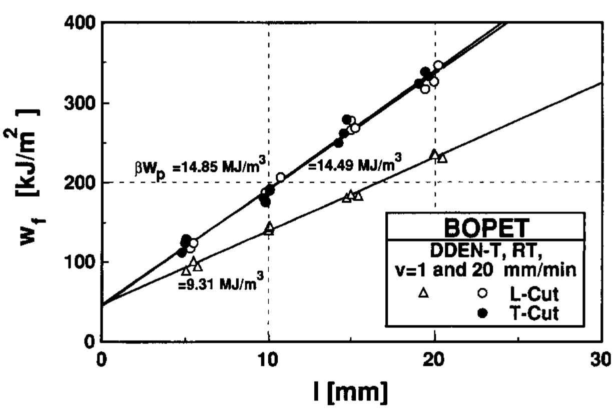

Figure 2 Total specific work of fracture (w_f) *versus* ligament length (l) from the DDEN-T specimens tested in L- and T-directions at $v = 20$ and 1 mm min^{-1}

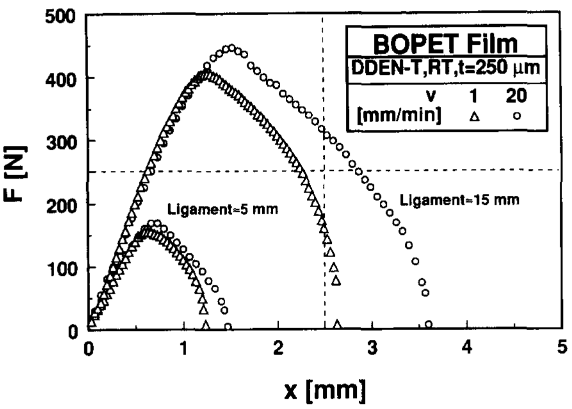

Figure 3 Comparison of the F–x curves of DDEN-T specimens of BOPET at similar ligament lengths ($l = 5$ and 15 mm) at $v = 20$ and 1 mm min^{-1}

($w_e = 53.6\,\mathrm{kJ\,m^{-2}}$)[5] and Hashemi ($w_e = 62.5\ \mathrm{kJ\,m^{-2}}$)[9] using the same specimen (i.e. DDEN-T) configuration. The PET studied by Chang and Williams[5] was an amorphous cast film of 180 μm thickness having a yield strength (σ_y) of 40 MPa and Young's modulus (E) of 2.3 GPa. On the other hand, the 250 μm thick PET film studied by Hashemi[9] was characterized by $\sigma_y = 85$ MPa and $E = 3.5$ GPa. The latter values indicate a semi-crystalline PET film that might have been oriented as well (unfortunately no further details were given by the author). BOPET in our case exhibited the following mechanical parameters (measured in both L- and T-directions): $\sigma_y = 103$ MPa and $E = 5.78$ GPa. These values were independent of the crosshead speed over a rather broad range; $v = 1$ to 100 mm min^{-1}.

The above PET samples can be classified with respect to their essential work of fracture as follows: amorphous < crystalline (possibly oriented) > crystalline (strongly oriented and filled). This ranking reflects that w_e is indeed affected by the microstructure of the semicrystalline PET. In addition, this ranking seems to support the statement of the author[6] that resistance to fracture usually goes through a maximum, as a function of crystallinity and crystalline orientation, in semicrystalline polymers. It should be kept in mind that this comparison is allowed only if the molecular characteristics of the PETs studied are similar (this can be supposed, since both the supplier and processing technique of the films were the same).

The validity range of the essential work approach is generally given by[6,9–11]

$$(3\text{–}5)t \leq l \leq \min\left(\frac{B}{3} \quad \text{or} \quad 2r_p\right) \tag{4}$$

where B is the width of the specimen (35 mm) and $2r_p$ is the size of the plastic zone:

$$2r_p = \frac{1}{\pi} \cdot \frac{Ew_e}{\sigma_y^2} \tag{5}$$

The plastic zone calculated by inserting the aforementioned mechanical parameters yielded $2r_p \approx 25$ mm. This size is much larger than that of the alternative criterion: $B/3 \approx 12$ mm. Based on the self-similarity of the F–x curves in the ligament range studied, which yielded a high correlation coefficient ($r \geq 0.990$) for the linear regression according to equation (3), it was assumed that the $B/3$ criterion is too conservative in this case.

Plastic work and related zone

w_p was determined by different approaches both in direct and indirect ways.

For assessing the plastic work dissipated per unit volume, uniaxial tensile tests on dumb-bells (*Figure 1*) were performed following the recommendations of the ESIS group[7]. The total energy up to failure, calculated from the area beneath the F–x curves, was divided by the necked region observed (thickness × width × initial length of the necked region $= 0.25 \times 10 \times 94\,\mathrm{mm^3}$). By this approach, $w_p = 49.2\,\mathrm{MJ\,m^{-3}}$ was received independent of the crosshead speed. Inserting this value into the slopes (i.e. βw_p) of the w_f *versus* l lines in *Figure 2*, $\beta \approx 0.30$ and 0.19 could be deduced for $v = 20$ and 1 mm min^{-1}, respectively.

Attention was paid also to the estimation of β by considering the IT pictures taken from the plastic zone during loading. *Figure 4* clearly shows that crack growth started before the ligament was completely yielded, at least for DDEN-T specimens with $l \approx 20$ mm. Based on the IT pictures in *Figure 4b* one can recognise that yielding along the full ligament takes place at position B of the related F–x curve (*Figure 4a*). Comparing the distance between the notch roots in the IT frames A and B (*Figure 4b*), the ligament reduction becomes obvious. This means that crack growth preceded the ligament yielding process. This scenario dominated the fracture of all DDEN-T specimens except those of the smallest ligament length ($l \approx 5$ mm). In their case crack growth started after full ligament yielding. An analogous fracture mode was observed for the PET film studied by Hashemi[9].

The series of IT pictures in *Figure 4b* portray the development of the temperature field in the ligament area during loading. The temperature rises by about 4°C in the ligament before final fracture, even when $v = 1$ mm min^{-1}. The cursor points 1 to 4 on the IT frames were positioned as follows: 1—far away from the ligament area and thus showing the reference temperature; 2 and 3—in the crack tip zones; and 4—in the mid ligament range. The IT frames in *Figure 4b* show that after full yielding and thus development of the plastic zone (see pictures B and C), crack growth accelerates. This crack acceleration, reflected also by the form of the F–x curve (*Figure 4a*), results in hot spots on both sides of the ligament (frames D and E) which travel towards one another until final fracture takes place in the mid ligament range (IT frame F in *Figure 4b*).

Figure 4b indicates further that the shape of the plastic zone can be approached by an ellipse (frames C to F in *Figure 4b*). The full height (h) of the plastic zone (by considering both halves of the DDEN-T specimens) just before fracture was read from the videotaped IT sequence and plotted against l. This resulted in a linear regression (*Figure 5*), the slope of which served for the determination of β (by accepting an elliptical shape for the plastic zone)[7]:

$$\beta = \frac{\pi}{4} \cdot \frac{h}{l} \tag{6}$$

In this way $\beta \approx 0.31$ and 0.28 were found for $v = 20$ and 1 mm min^{-1}, respectively. Considering the above

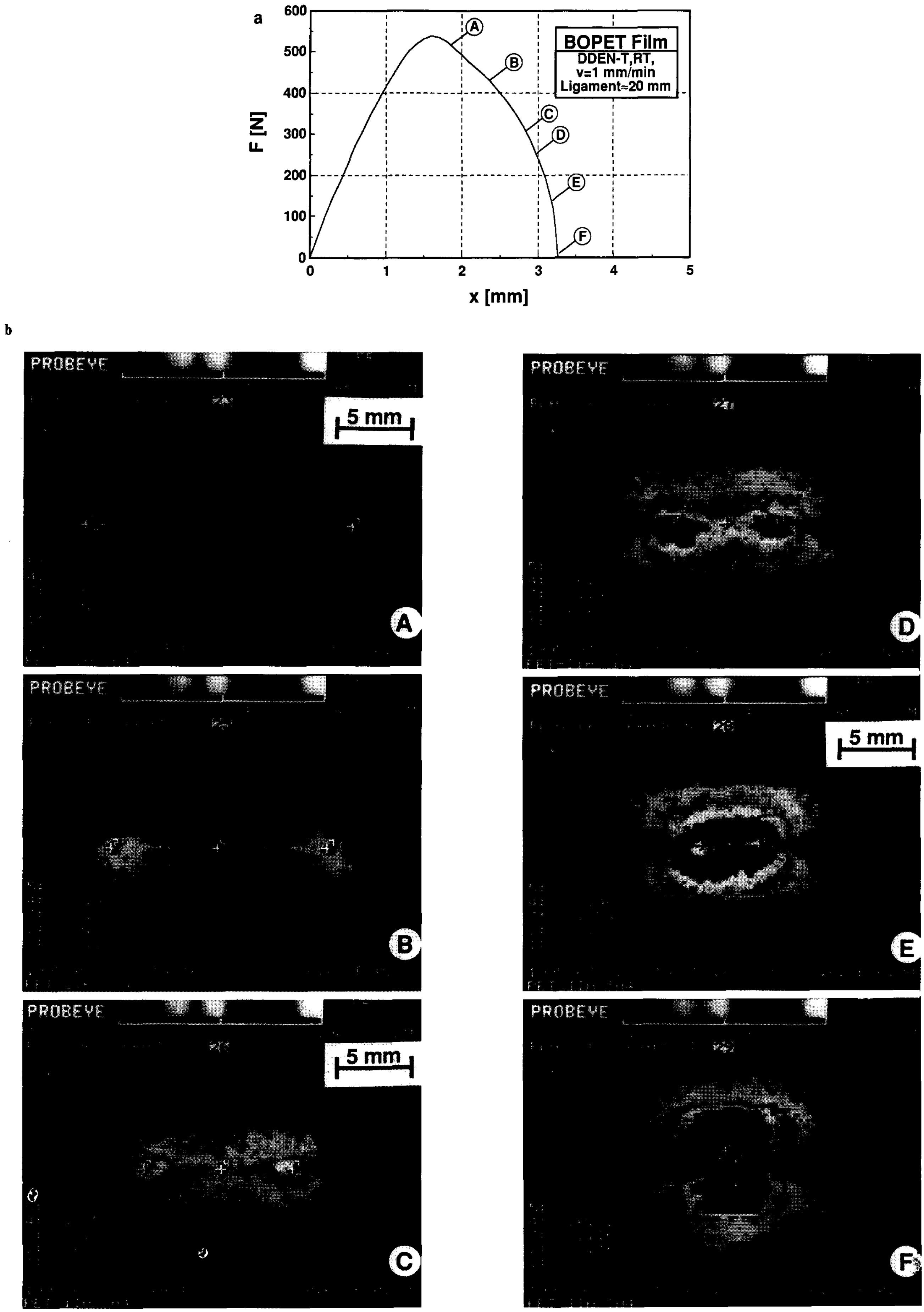

Figure 4 F–x curve of a DDEN-T specimen with l = 20 mm (a) along with a series of IT frames taken during loading (b). Note: the positions at which the IT frames were taken are indicated in the F–x curve in *Figure 4a*

β-values for the related plastic work, from the slopes of the regression lines in *Figure 2*, the values $w_p = 46.8$ and $34.3\,\mathrm{MJ\,m^{-3}}$ were calculated for the higher and lower crosshead speed, respectively. It is noteworthy that the former value is very close to that derived from the tensile tests on dumb-bells (see *Table 1*).

w_p can be determined directly from trouser tests as well[1,8,9]:

$$w_p = \frac{F}{t^2} \tag{7}$$

where F is the equilibrium (constant) load. This approach yielded w_p values in the range of 75 to $78\,\mathrm{MJ\,m^{-3}}$, independent of the crosshead speed used. Substituting these values into the slopes of *Figure 2*, $\beta \approx 0.19$ can be computed for the shape parameter of the plastic zone (*Table 1*).

It should be noted that the trouser test can also be used for the direct determination of w_e[1,12]:

$$w_e = \frac{2F}{t} (= 2w_p t) \tag{8}$$

Based on equation (8), $w_e = 37\text{–}39\,\mathrm{kJ\,m^{-2}}$ could be determined; this range is far below that found experimentally on DDEN-T specimens ($w_e = 45\text{–}47\,\mathrm{kJ\,m^{-2}}$). There is no controversy with this finding. It is well known that the resistance to fracture under mode I can be substantially enhanced by monoaxial (or biaxial) orientation. This improvement occurs, however, at the cost of the resistance to out-of-plane type (transverse, mode III) deformation when the crack growth follows the orientation direction. This is due to the molecular orientation and related supermolecular arrangement being affected by the drawing process[13]. A possible rationale behind the finding that w_e (mode III) < w_e (mode I) may reside in the anisotropic response of the tie molecules which connect lamellar and fibrillar entities. They can be more easily deformed in-plane by lamellar (rotation, defolding and breaking-up) and fibrillar rearrangements than under out-of-plane type loading. In the latter case, severe stress concentration is prevailing that hampers an adequate microstructural response of the polymer under the mode III loading situation.

The w_e and w_p values, determined directly from the trouser tests, do not match those from the DDEN-T specimens. It is reasonable to accept that the difference between them increases with greater in-plane orientation of the film. Thus, w_p values derived from the trouser test cannot be used to estimate the β parameter for different test geometries under mode I deformation. This claim is at odds with the report of Hashemi[9].

It was of interest to ascertain whether or not the necking is associated with strain-induced crystallization. Comparing both the d.s.c. traces (*Figure 6*) and the

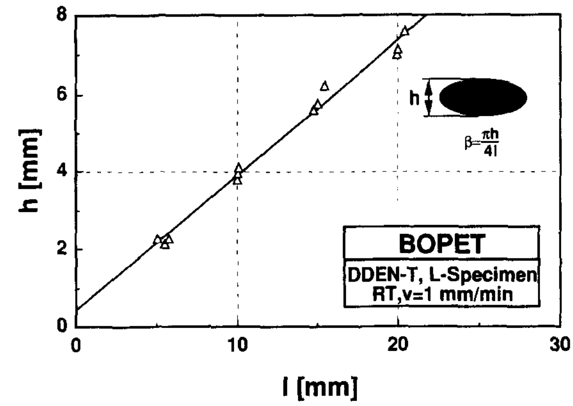

Figure 5 Total height of the plastic zone (h) determined by IT *versus* ligament length (l) for DDEN-T specimens

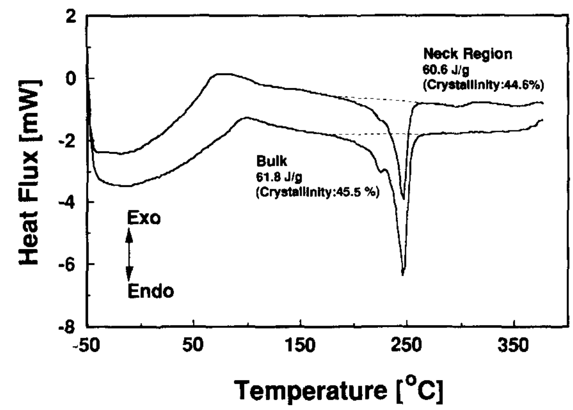

Figure 6 D.s.c. traces of samples taken from the bulk and necked region of a failed DDEN-T specimen

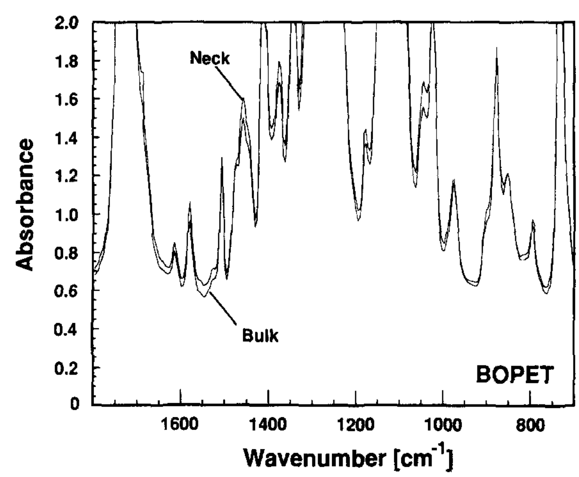

Figure 7 Comparison of the *FT*i.r. spectra of samples taken from the bulk and necked area

Table 1 Comparison of the non-essential or plastic work (w_p) and shape parameter of the plastic zone (β) determined by different approaches; arrow indicates the calculation route

<table>
<tr><th rowspan="2">Method</th><th colspan="2">w_p ($\mathrm{MJ\,m^{-3}}$)</th><th rowspan="2"></th><th colspan="2">β</th></tr>
<tr><th>$v = 20\,\mathrm{mm\,min^{-1}}$</th><th>$v = 1\,\mathrm{mm\,min^{-1}}$</th><th>$v = 20\,\mathrm{mm\,min^{-1}}$</th><th>$v = 1\,\mathrm{mm\,min^{-1}}$</th></tr>
<tr><td>Tensile test on dumb-bell specimens</td><td colspan="2">49.2</td><td>→</td><td>0.30</td><td>0.19</td></tr>
<tr><td>Infra-red thermography (IT)</td><td>47.8</td><td>34.3</td><td>→</td><td>0.31</td><td>0.28</td></tr>
<tr><td>Tearing test on trouser-type specimens</td><td colspan="2">76.5</td><td>→</td><td>0.19</td><td>0.19</td></tr>
</table>

FT-i.r. spectra (*Figure 7*) of samples taken from the bulk and necked region of the DDEN-T specimens, no difference could be revealed. The crystallinity of the samples was $\sim 45\%$ (accepting $136\,J\,g^{-1}$ melt enthalpy for the fully crystalline PET[14]) and did not change with the sampling position. Keeping in mind that the *FT*i.r. technique reflects conformational changes caused by drawing very sensitivity[15], the phenomenon of strain-induced crystallization can be excluded in our case.

CONCLUSIONS

The fracture toughness of biaxial-oriented, filled poly-(ethylene terephthalate) (BOPET) was studied by the essential work of fracture method using deeply double-edge notched tensile (DDEN-T) specimens. The essential work of fracture (w_e) was independent of the crosshead speed and seemed to be a material parameter, at least for the given film thickness (t). The non-essential or plastic work of fracture (βw_p, where β is the shape parameter or the plastic zone) turned out to be rate-dependent. Direct evidence for a change in the plastic zone was delivered only by infra-red thermography (IT). The IT technique also allowed the sequence of failure to be clarified. At low ligament length necking of the whole ligament preceded the initiation of the crack growth, whereas at higher ligaments necking and crack growth occurred simultaneously in the post-maximum range. Among the different approaches applied to determine w_p explicitly, IT seemed to be the most promising. This, in turn, substantiates the suggestion to use IT for the direct determination of PYFM parameters, such as the *J*-integral[16]. The out-of-plane type deformation (mode III) in the trouser test resulted in markedly lower w_e and higher w_p values than those obtained by in-plane (mode I) tests on DDEN-T specimens. The difference in the mean w_e values from mode I and mode III tests was attributed to a microstructure-related strong mechanical anisotropy of the BOPET film. This finding indicates that caution should be exercised when estimating fracture parameters and related terms from tests performed under mode I and mode III.

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