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Thermal effects on fracture of biaxial-oriented poly(ethylene terephthalate) (BOPET) film

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Abstract The effect of temperature on the fracture behaviour of biaxial-oriented poly(ethylene terephthalate) (BOPET) film was studied using the Essential Work of Fracture (EWF) approach. Fracture tests were performed over the temperature range +25 to +160 °C at the speed of 5 mm/min using double edge notched tension (DENT) specimens. The length of the specimens was either along the machine direction (MD) (0°), transverse direction (TD) (90°) or at 45° to either MD or TD. Ductile tearing of the ligament region was noted over the entire temperature range in all three directions. A linear relationship was found between the specific total work of fracture and the ligament length at all test temperatures. Values of the specific essential work of fracture (w_e) in the MD and TD were similar and smaller than in the 45° direction. Within temperature range 25–140 °C, w_e showed little variation if any with respect to temperature. As expected, the Specific Non-Essential Work of Fracture (βw_p) was temperature dependent. This parameter increased with increasing temperature and reached a maximum around the glass transition temperature of BOPET ($T_g \approx 80$ °C). The values of the maxima are respectively 16.15, 20.38 and 17.8 MJm^{-3} for the 0°, 45° and 90°.

Introduction

It is well established that fracture behaviour of polymers can be characterized either by the critical value of the stress intensity factor, K_c or by the critical value of the J-integral, J_c. Both parameters are generally used in determining fracture toughness under plane strain conditions which is independent of geometry and the dimensions of the test piece. To achieve plane-strain conditions, the thickness of the test specimen must be greater than a certain value prescribed by the ASTM [1, 2]. For high toughness and low yield strength polymers the required thickness could be quite large and difficult to manufacture. Many polymers, such as poly(ethylene terephthalate) are widely used in packaging in thin-film forms with sub-millimeter thickness. As it is impossible to test such films using the standardized geometries such as single edge notched bend (SENB) or compact tension (CTEN), any measured value of K_c or J_c is most likely to be invalid, simply because the thickness requirement cannot be met and therefore the stress state in the specimen becomes one of plane-stress rather than plane-strain or combination of the two modes (i.e., mixed mode).

To characterize fracture toughness of polymeric films, many authors (e.g., [3–27]) have used the method of the Essential Work of Fracture (EWF) as originally proposed by Broberg [28] in order to characterize the fracture behaviour of ductile materials that fail under plane-stress conditions. The EWF approach offers an attractive means for separating the energy spent in the fracture process zone from the work spent in the plastic deformation zone that surrounds the fracture process zone. The former is referred to as ''Essential Work of Fracture'' and the latter as ''Non-Essential Work of Fracture''. The EWF method seeks to determine a fracture parameter called ''Specific Essential Work of Fracture, w_e'' which is the measure of the material toughness under plane-stress conditions.

In the present work, a biaxial-oriented poly(ethylene terephthalate) (BOPET) film was tested over the tempera-

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ture range 25–160 °C using deep notched double edge notched geometry with the purpose of evaluating the influence of temperature and orientation on w_e.

Essential work of fracture (EWF)

The methodology

Broberg [28] stated that when ultimate failure of the pre-notched specimens is preceded by extensive yielding and slow crack growth, a toughness parameter called specific essential work of fracture, w_e, may be evaluated. According to Broberg, the nonelastic region at the crack tip may be divided into two regions as shown in Fig. 1: An inner fracture process zone (IFPZ) and an outer plastic deformation zone (OPDZ). The inner fracture process zone is where fracture actually takes place and an outer plastics deformation zone is where various types of deformations such as shear yielding and microvoiding may be operating. The energy term associated with IFPZ is termed the ''Essential Work of Fracture (W_e)'' and that associated with OPDZ is termed the ''Non-Essential Work of Fracture (W_p)''. The total work of fracture W_f which is given by the area under load-displacement (P-δ) curve of a notched specimen is then written as:

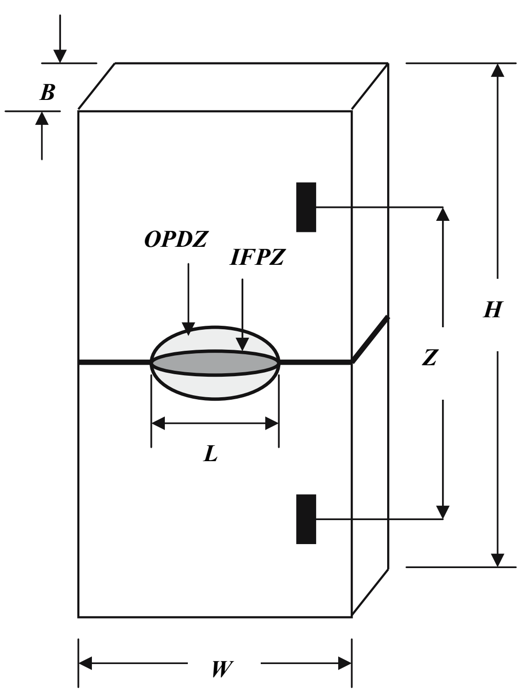

Fig. 1 Inner fracture process zone (IFPZ) and outer plastic deformation zone (OPDZ). (B = Specimen thickness, W = Specimen width, Z = Gauge length and H = Specimen length)

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

The term W_e is the work expended in the IFPZ to form a neck and its subsequent tearing. Physically, W_e is a pure crack resistance parameter and is essentially a surface energy term whose value is proportional to the ligament area (LB), i.e.,

$$W_e = w_e\, BL \tag{2}$$

The term w_e is termed the ''Specific Essential Work of Fracture'' and is considered to be a material constant for a given constant.

The term W_p in Eq. 1 is the work dissipated in the OPDZ and is assumed to be proportional to the volume of the yielded zone (BL^2), i.e.,

$$W_p = w_p \beta BL^2 \tag{3}$$

The term w_p is termed the ''Non-Specific Essential Work of Fracture''. The parameter β is a proportionality constant or shape factor associated with the volume of the plastic deformation zone whose value depends on the geometry of the specimen and the crack.

Introducing Eqs. 2 and 3 into Eq. 1 gives:

$$w_f = \frac{W_f}{LB} = w_e + \beta w_p L \tag{4}$$

where w_f is termed the ''Specific Total Work of Fracture''.

According to Eq. 4, the plot of w_f as a function of L should be linear, whose intercept with the w_f axis gives w_e, and whose slope gives βw_p. Thus, the EWF method involves testing specimens with different ligament lengths, measuring W_f for each (area under the load-displacement diagram), plotting the w_f–L diagram and performing linear regression analysis for determining w_e and βw_p.

Specimen size criteria for valid EWF

It has been recommended that for w_f and L be linearly related as required by Eq. 4, the length of the ligament region should satisfy the following lower and upper bound threshold values [29];

$$\begin{aligned} L_{min} &= 3B \text{ to } 5B \\ L_{max} &= \min(W/3,\ 2R_p) \end{aligned} \tag{5}$$

where R_p is the radius of the plastic zone at the crack tip.

The above limitations are imposed upon ligament length to ensure the following:

(i) Complete yielding of the ligament region occurs prior to crack growth, hence the condition

$L_{max} = 2R_p$. This condition ensures the proportionality of W_p and L^2 as required by Eq. 3 is maintained.

(ii) Size of the plastic zone is not disturbed by the boundaries of the test specimen (i.e., edge effects), hence the requirement $L_{max} = W/3$.

(iii) State of pure plane-stress is developed in the ligament region, hence the requirement $L_{min} = 3B–5B$. This ensures that w_e and w_p are independent of L. For $L < L_{min}$, the state of stress in the ligament region becomes one of mixed-mode, in which case it would have both plane-stress and plane-strain characteristics. As a consequence of this, the relationship between w_f and L is no longer linear as required by Eq. 4.

To verify (iii), it is customary to plot values of net-section stress at maximum load, σ_n, versus ligament length. According to Hill [30] σ_n is independent of ligament length in the plane-stress region (having value of $1.15\sigma_y$ for double edge notched tension specimen), but increases with shortening of the ligament region in the mixed mode region. Although the transition from one region to another is a gradual one, it is nonetheless possible to make a reasonable estimate of L_{min} at which this transition takes place and discard values of w_f for specimens with $L \leq L_{min}$ when performing the regression analysis on w_f versus L.

Experimental details

Material

The material studied in this work was a Du Pont biaxially oriented PET film supplied in the form of DIN-A4 sheets of nominal thickness 0.125 mm. The film was translucent white and was oriented in two directions—the direction of extrusion (machine direction, MD) and perpendicular to the machine direction (transverse direction, TD). The glass transition temperature of the PET material is 80 °C.

Tensile tests

Determination of tensile yield stress and modulus were carried out between 25 °C and 160 °C using dumbbell-shaped specimens having constant width of 4 mm in the gauge length region. The dumbbell specimens were punched from the sheets such that the long axis of the specimens was either along MD (0° direction), TD (90° direction) or at 45° to either MD or TD. At least five specimens from each direction were tested at a given temperature. The specimens were tensile tested in an Instron testing machine at a constant crosshead displacement rate of 5 mm/min using pneumatic clamps with an initial separation of 40 mm. The load-displacement curve (P-δ for each specimen was recorded using a computer data logger.

Fracture tests

EWF measurements were carried out between 25 °C and 160 °C at a crosshead speed of 5 mm/min. Double edge notched tension (DENT) specimens were prepared by first cutting the sheets into rectangular coupons having a constant width (W) of 35 mm and overall length (H) of 105 mm (with a length between grips of $Z = 70$ mm) as shown in Fig. 1. The long axis of the coupons was either along MD (0° direction), along TD (90° direction) or at 45° to either MD or TD. The initial notches were made perpendicular to the traction direction using a fresh razor blade. At least 15 specimens with varying ligament lengths between 4 mm and 27 mm were prepared for determining a single w_e value. The ligament lengths were measured before the test using a traveling microscope. The load-displacement curve (P-δ for each specimen was recorded using a computer data logger, and the total work of fracture (W_f) was obtained by integrating the area under the curve. The validity of the data was checked by plotting the net-section stress at maximum load (σ_n) versus ligament length, and observing as to whether the values were consistent with the Hill's [30] prediction of $\sigma_n = 1.15\sigma_y$ in plane-stress.

Results and discussion

Deformation characteristics

Figure 2 shows typical tensile load-displacement (P-δ curves obtained in this study in the 0°, 45° and 90° directions. It can be seen from the curves that the direction had no significant effect upon the extensional stiffness and

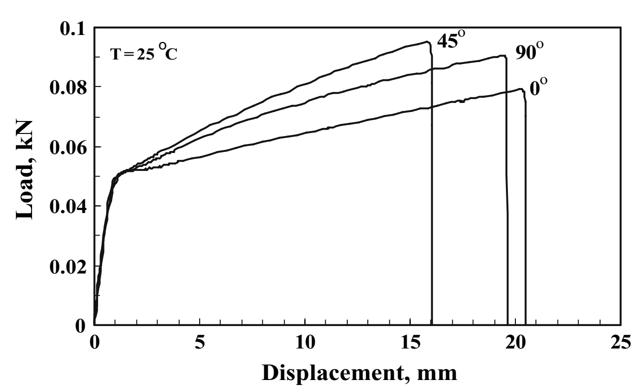

Fig. 2 Typical tensile load-displacement (P-δ) curves for BOPET showing the effect of specimen orientation on tensile deformation at 25 °C

tensile yield stress of BOPET. There is however obvious differences in deformation in the post yield regime. As illustrated by the P-δ curves in Fig. 2, tensile stress and the rate at which it increases with strain (i.e., strain hardening) is highest in the 45° direction and lowest in the 0° direction; opposite trend can be seen for the elongation at break. This behaviour, which was consistently observed over the entire temperature range, further emphasizes the importance of film anisotropy and suggests that among the three directions, the best resistance to traction is in the 45° direction and the worst is in the 0° direction.

The effect of temperature on P-δ curve is shown in Fig. 3 where it can be seen that whilst the curves obtained at $T \leq 80$ °C show a yield point, those obtained at higher temperatures do not. For this reason, the load at 2% offset strain was arbitrary chosen for determination of the tensile yield strength. The temperature dependence of the tensile yield strength (σ_y) and modulus (E) in the 0°, 45° and 90° directions is shown in Fig. 4. As expected, values of σ_y and E decrease with increasing temperature owing to visco-elasticity of the material. It is note worthy that over the entire temperature range, neither σ_y nor E show any significant variation with respect to the direction in which the specimens were punched from the sheets.

Crack growth characteristics

The P-δ curves for DENT specimens examples of which are shown in Fig. 5 as a function of ligament length indicated that failure of BOPET specimens was always by ductile tearing of the ligament region. The notable feature of the curves obtained in this study over the entire temperature range was their geometrical similarity, which is an essential pre-requisite for EWF testing. The specimen direction had no major influence upon the general behaviour of the P-δ curves.

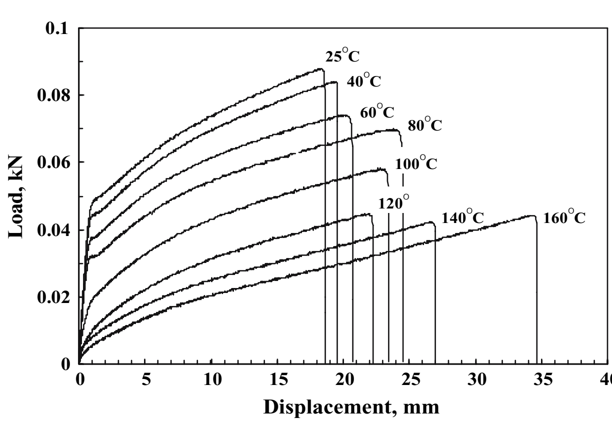

Fig. 3 Typical tensile load-displacement (P-δ curves for BOPET showing the effect of temperature on tensile deformation (90° direction)

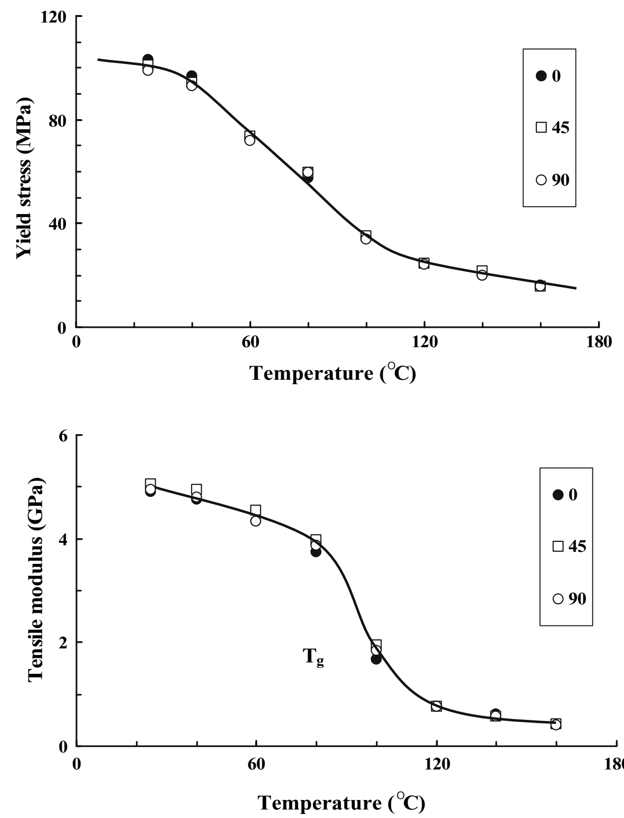

Fig. 4 Temperature dependence of the tensile yield strength and modulus for BOPET in the three directions

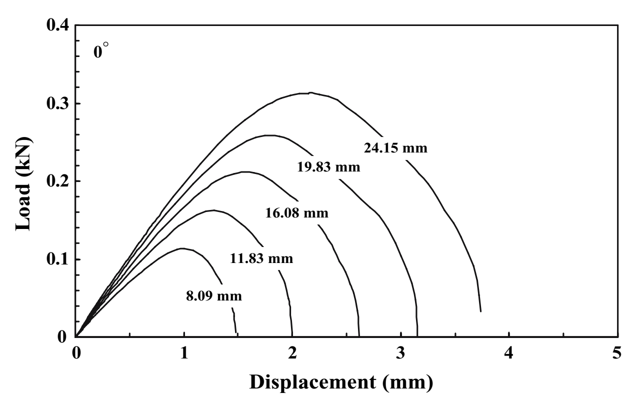

Fig. 5 Typical DENT load-displacement curves at various ligament lengths at 25 °C for the 0° direction

Figure 6 illustrates typical P-δ curves at various temperatures for a constant ligament length ($L \approx 16$ mm). It can be seen that whilst maximum load decreases with increasing temperature, the extension at break and the rate at which load after maximum diminishes to zero depends largely as to whether the test temperature was higher or lower than the T_g of 80 °C. Evidently, load after maximum diminishes to zero at a faster rate when T exceeds T_g. Moreover, when the test temperature exceeded 100 °C,

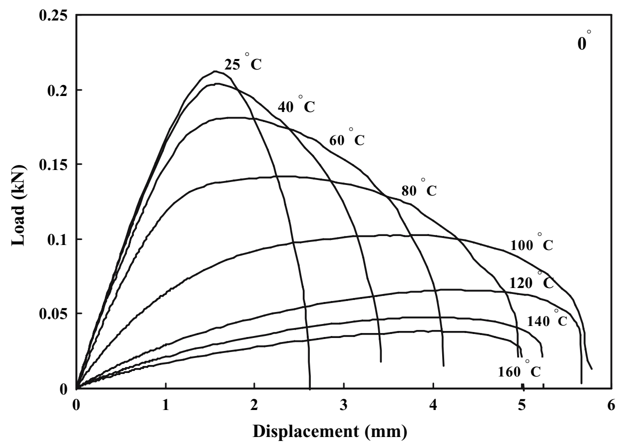

Fig. 6 Typical load-displacement curves for DENT specimens at various temperatures for a fixed ligament length of 16 mm

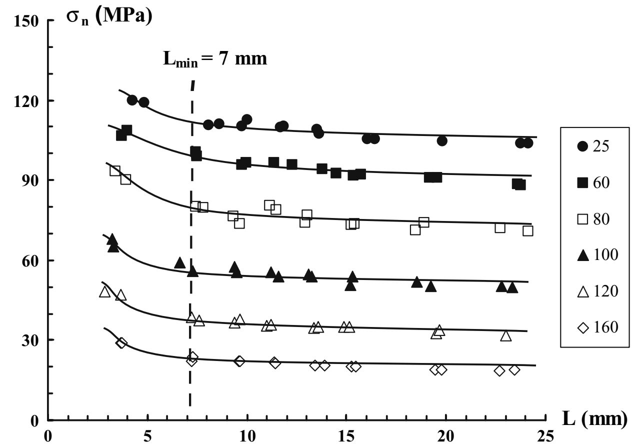

Fig. 7 Plots of net-section stress at maximum load versus ligament length at some selected test temperatures in the 0° direction

extension at break decreased with increasing temperature. These observations imply that the material resistance to crack propagation is temperature dependent with maximum resistance occurring at or near the T_g.

It is worth noting that although propagation of the crack was visually detectable in all the specimens tested here, crack tip yielding could not be observed since the material in the plastic deformation zone had the same refractive index as the bulk material. For this reason, it cannot be confirmed (i) if the ligament regions were fully yielded prior to crack growth and (ii) if at maximum load full yielding of the ligament regions was completed. However, since propagation of the crack in all the specimens began before maximum load was reached, the decrease in load after maximum was attributed mainly to the loss of resistance caused by the reduction in the ligament length region.

Analysis of the net-section stress

In order to verify the validity of the EWF data, values of net-section stress at maximum load σ_n were plotted against ligament length for the 0°, 45° and 90° directions as a function of temperature. Plots indicated that whilst σ_n was not affected significantly by the orientation, it decreased with increasing temperature thus reflecting the temperature dependency of the tensile yield stress. The effect of temperature on σ_n versus L can be seen from the examples shown in Fig. 7.

As for the effect of ligament length on σ_n, it can be seen from Fig. 7 that σ_n decreases with increasing L with a clear evidence of a sharp increase in σ_n at the small ligament lengths which is suggestive of some mode mixity for ligament lengths shorter than 7 mm.

Although the observed trend of decreasing σ_n with L is common to many EWF studies, it is worth commenting on two aspects of the data, one in relation to the pre-requisite L_{min} = 3*B*–5*B* and the other in relation to the theoretical plane-stress value of $1.15\sigma_y$.

Firstly, we acknowledge that defining a precise value for L_{min} is difficult, however, based on σ_n versus L plots (see Fig. 7), value of 7 mm appears to be a reasonable estimate for L_{min} for the entire temperature range under consideration. This value suggests that stress state transition in 0.125 mm thick BOPET occurs at L_{min}/B ratio of approximately 56. Clearly, whatever the selection criteria for L_{min}, the recommended transitional ratio of 3–5 (i.e., 0.375–0.625 for B of 0.125 mm) would not be adequate to guarantee plane-stress conditions. This observation and the fact that studies on polymeric films have yielded a wide-ranging values of L_{min}/B, further supports the view that this ratio depends on the material and therefore does not have a universal value as originally perceived.

Secondly, the theoretical plane-stress value of $1.15\sigma_y$ is rarely achieved in this study. Indeed, values of σ_n obtained in this study were generally greater than the theoretical value and the difference between the two widened with increasing temperature. This observation is also common to several studies and indeed it is nowadays believed that EWF results should still be valid so long as σ_n has reached some steady state value, even though the value may not be $1.15\sigma_y$.

Specific essential and non-essential work of fracture

Figures 8–10 show plots of the specific work of fracture, w_f, versus ligament length, L, in the 0°, 45° and 90° directions between 25 °C and 160 °C. It can be seen that in all cases w_f increases with increasing L in a linear manner for ligament lengths in the range 7–25 mm. However, for ligament lengths less than 7 mm, a nonlinear trend is obtained owing to lower than expected values of w_f. This

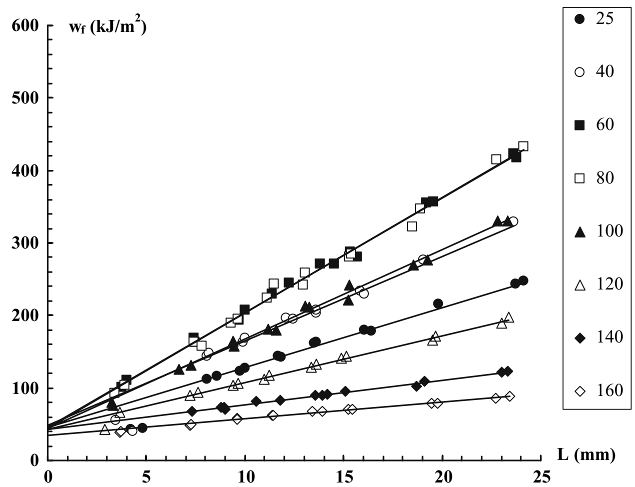

Fig. 8 Specific work of fracture versus ligament length for the 0° direction at various temperatures

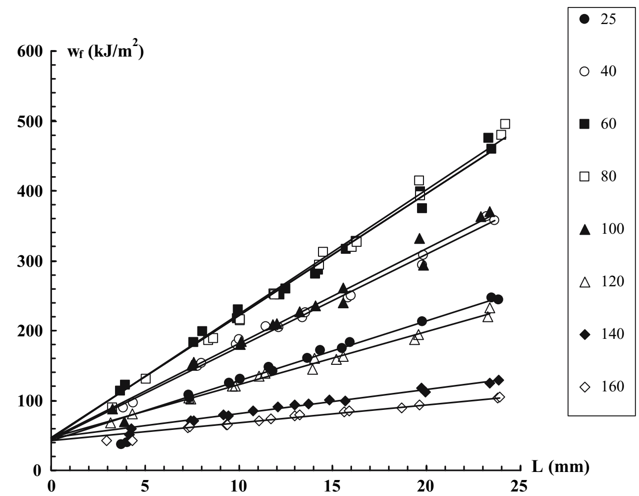

Fig. 10 Specific work of fracture versus ligament length for the 90° direction at various temperatures

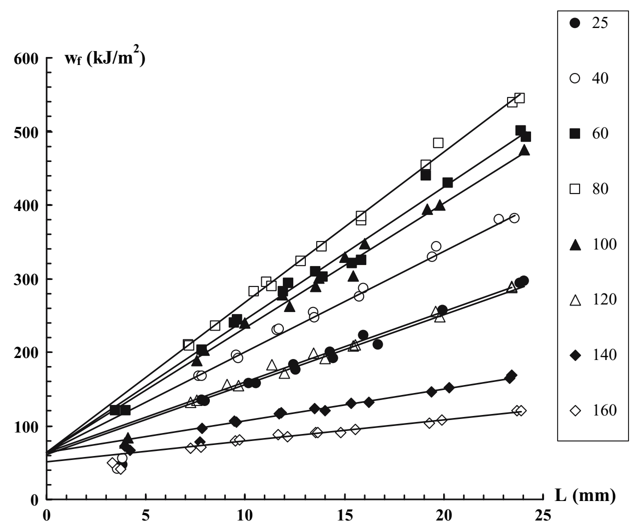

Fig. 9 Specific work of fracture versus ligament length for the 45° direction at various temperatures

observation indicates mode mixity in the ligament region as verified by σ_n versus L plots.

To determine the plane-stress values of the specific essential and non-essential work of fracture as a function of temperature, the w_f values obtained for ligament lengths in the range 7–25 mm were considered for linear extrapolation to $L = 0$, since only within this range W_p was proportional to L^2, as it is required by Eq. 4. The w_e and βw_p values obtained in this way are given in Table 1 along with the regression coefficients (r^2) which are given in the parentheses.

It can be seen from the values in Table 1 that w_e is not affected significantly by the temperature at least between 25 °C and 140 °C. It is also notable that values of w_e in the 0° (MD) and the 90° (TD) directions are markedly similar, both being consistently smaller than in the 45° direction. It is worth pointing out, that Karger-Kocsis [7] reported MD and TD values of w_e in the range 45–47 kJ/m^2 for 0.25 mm thick chalk-filled BOPET film at 23 °C and Maspoch et al. [17] reported MD values in the range 46–51 kJ/m^2 and TD values in the range 62–67 kJ/m^2 for 0.05 to 0.25 mm thick BOPET films at 23 °C. Clearly, whilst Karger-Kocsis [7] study shows no orientation effect on w_e, Maspoch et al. [17] study indicates a clear and a strong orientation effect with TD values being 25–30% higher than MD values. The MD and TD values of w_e obtained in this study at 25 °C are in good agreement with the values reported by Karger-Kocsis [7].

As for βw_p, it can be seen from the tabulated values in Table 1 and their graphical representation in Fig. 11 that this parameter increases rapidly with increasing temperature and reaches a maximum at around the glass transition temperature of BOPET (≈ 80 °C). βw_p was highest in the 45° direction and lowest in the 0° direction. This observation is consistent with the tensile load-displacement curves of Fig. 2 showing a greater strain hardening effect in the 45° direction than in the 0° direction.

As stated earlier, purpose of the requirement $L_{max} \leq W/3$ is to ensure the size of plastic zone is not disturbed by the boundaries of the test specimen (edge effects). This requirement suggests an upper bound ligament length value of approximately 11.67 mm for the specimen width of 35 mm. The requirement $L_{max} \leq 2R_p$ ensures complete yielding of the ligament region takes place prior to crack growth. Some typical values of $2R_p$ were calculated using the relationship $R_p \frac{1}{2\pi}\left(\frac{Ew_e}{\sigma_y^2}\right)$. It can be deduced from the values given in Table 2 and the w_f versus L plots in Fig. 8 that neither $W/3$ nor $2R_p$ has any

Table 1 Specific essential and non-essential works of fracture in the 0° direction

Direction	0°		45°		90°	
EWF parameters	w_e (kJ/m^2)	βw_p (MJ/m^3)	w_e (kJ/m^2)	βw_p (MJ/m^3)	w_e (kJ/m^2)	βw_p (MJ/m^3)
25 °C	45.26 ± 2.49 (0.996)	8.26 ± 0.16	60.75 ± 4.50 (0.991)	9.50 ± 0.29	42.75 ± 2.40 (0.996)	8.60 ± 0.16
40 °C	47.87 ± 3.90 (0.995)	11.80 ± 0.27	64.08 ± 4.29 (0.995)	13.77 ± 0.28	43.65 ± 4.38 (0.993)	1301 ± 0.30
60 °C	46.25 ± 3.40 (0.997)	15.79 ± 0.23	63.24 ± 12.84 (0.978)	18.05 ± 0.80	47.29 ± 6.39 (0.991)	17.04 ± 0.44
80 °C	42.11 ± 6.95 (0.991)	16.15 ± 0.46	63.60 ± 5.13 (0.997)	20.38 ± 0.34	46.00 ± 5.13 (0.992)	17.80 ± 0.34
100 °C	43.57 ± 4.44 (0.993)	12.36 ± 0.30	64.51 ± 9.24 (0.986)	17.03 ± 0.61	45.20 ± 8.36 (0.981)	13.67 ± 0.54
120 °C	44.16 ± 1.87 (0.996)	6.41 ± 0.12	63.95 ± 4.03 (0.991)	9.56 ± 0.26	46.95 ± 2.77 (0.991)	7.59 ± 0.19
140 °C	42.89 ± 1.36 (0.992)	3.40 ± 0.091	63.74 ± 1.83 (0.992)	4.36 ± 0.12	46.92 ± 1.41 (0.991)	3.44 ± 0.095
160 °C	34.36 ± 1.19 (0.986)	2.30 ± 0.078	50.61 ± 1.54 (0.986)	2.91 ± 0.099	42.60 ± 0.92 (0.993)	2.59 ± 0.061

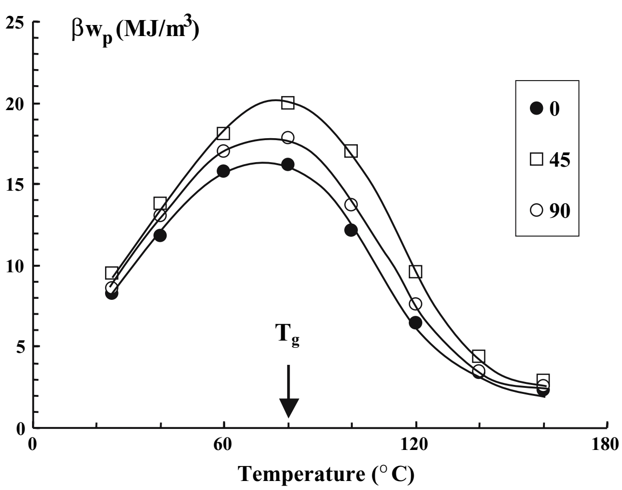

Fig. 11 Specific non-essential work of fracture versus temperatures in the three directions

relevance here since the proportionality of w_p with L^2 is maintained well beyond L_{max}. This begs the question as to whether any correlation exists between L_{max} and the plastic zone size.

Table 2 Size of plastic zone in the 0° direction as a function of temperature.

T (°C)	$2R_p$ (mm)
25	6.62
40	7.77
60	11.69
80	15.48
100	19.01
120	18.03
140	20.82
160	17.50

Conclusion

The effect of temperature on the specific essential and non-essential work of fracture of BOPET film was investigated between 25 °C and 160 °C using double edge notched tension specimens. The length of the specimens was either along the MD (0°), TD (90°) or at 45° to either MD or TD. It was noted, that the fracture of BOPET film was always by ductile tearing of the ligament region. However, due to absence of the stress whitening phenomenon at the crack tip, it was not possible to detect visually whether crack propagation occurred after or before full yielding of the ligament region. Nonetheless, a linear relationship was found between the specific total work of fracture and ligament length over the entire temperature range in all three directions (MD, TD and 45°).

It was found that w_e for the 0° and 90° directions were similar and smaller than in the 45° direction. The w_e for all three directions was found to be independent of temperature, at least between 25 °C and 140 °C.

As expected, βw_p was found to be dependent upon temperature, increasing with temperature and reaching a maximum at the glass transition temperature of BOPET. βw_p was also dependent upon orientation; the highest value was obtained in the 45° direction and lowest in the 0°direction.

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