The effect of temperature on the fracture of rubber modified polystyrene

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Brittle fractures were obtained in rubber modified polystyrene over the temperature range -120 to 20° C using the surface notch method. When compared with single edge notch data, a thickness effect was apparent and this was described in terms of plane stress and plane strain fracture toughness values. The plane strain value agreed closely with that of the polystyrene matrix indicating that the constrained region showed no toughening effect. The relaxation process of the rubber was apparent in the plane stress toughness.

1. Introduction

Polystyrene is one of the most important commercial polymers and in its rubber toughened forms (high impact polystyrene, HIPS, and acrylobutadienestyrene, ABS) is used widely in applications requiring substantial levels of toughness. The toughening process has been widely discussed (e.g. [1]) and may be described briefly as the rubber particles acting as initiation points for the production of energy absorbing crazes resulting in a considerable increase in the fracture toughness over the basic polymer. The visco-elastic properties of the rubber phase are considered to be a contributing factor in the toughening process [2]. This paper seeks to offer some clarification of these mechanisms by describing the toughness in fracture mechanics terms and using concepts developed with other polymers to elucidate the energy absorbing processes.

Previous work on both PMMA and polystyrene [3,4] has shown that the presence of a viscoelastic relaxation process around room temperature results in stable slow crack growth which elevates the fracture toughness of the material. The β process at 20° C is quite strong in PMMA leading to a substantial effect but in polystyrene it is less pronounced. By testing over a wide temperature range, the influence of the β transition in PMMA was further illustrated [5] so that when the instability fracture toughness, $K_{\rm C}$, was plotted as a function of temperature, a curve of $K_{\mathbf{C}}$ increasing with decreasing temperature was produced over the β transition range. Similar work on polycarbonate showed the same effect [6] but here there was evidence of a pronounced thickness effect which was absent in both PMMA and polystyrene. By using plane stress and plane strain fracture toughness values (K_{C2} and K_{C1}), it was possible to describe the data in the same terms as PMMA. Some fracture mechanics studies have been made on toughened polystyrene at 20° C [7] and because of the large degree of crazing, the specimens used (50 mm wide) underwent gross deformation and it was necessary to invoke the ductile failure criterion J_{C} to describe the data. Attempts to produce brittle failures for which K_{C} is an appropriate description were unsuccessful.

In the study reported here polystyrene and a rubber toughened grade were fracture tested over the temperature range -120 to 20° C. Thickness effects were apparent in the toughened material of a similar nature to polycarbonate and by the use of the surface notch technique it was possible to produce brittle failure over the whole temperature range.

2. Some theoretical concepts

The fracture toughness used in this work is calculated from the usual result:

$$K_{\rm C}^2 = Y^2 \sigma^2 a \tag{1}$$

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Figure 1 Geometry of surface notch specimens.

where Y^2 is a geometric correction parameter depending upon the specimen geometry. For a centrally notched infinite plate it is π and for a wide single edge notched (SEN) plate, 4. Some of the data reported here were obtained using SEN specimens and the appropriate function for Y^2 used (see [3-6]). Tests were also performed using the surface notch (SN) geometry which is shown in Fig. 1. A dead sharp flycutter is used to produce a notch in the form of a circular arc in the surface of the specimen. The expression for K_C must be corrected for the different crack front length to crack area ratio as compared with the usual through-thickness crack and this is done using:

where

$$Y^{2} = \frac{1.21\pi}{\phi^{2} - 0.212(\sigma/\sigma_{y})^{2}}$$
$$\phi = \int_{0}^{\pi/2} \left\{ 1 - \left(\frac{b^{2} - d^{2}}{b^{2}}\right) \sin^{2}\theta \right\}^{\frac{1}{2}} d\theta$$

 $K_{\mathbf{C}}^2 = Y^2 \sigma^2 a$

and σ_{y} = the tensile yield stress [9].

It will also be assumed here (see [6]) that the surface notch has an equivalent thickness H' given by a crack of uniform depth d and the same surface area. For the circular form with $b \ll R$ (the radius of curvature):

$$H' \simeq \frac{4}{3}b. \tag{3}$$

(2)

It should be noted that Equation 2 is deduced assuming a semi-elliptical form for which the result is $H' = \pi/2b$.

It has been found helpful previously [6] to assume that there are two fracture toughnesses for a polymer; one under plane stress conditions, K_{C2} , and one under plane strain, K_{C1} . The concept is well known in the metals field but here it is invoked to describe flat fracture or at least those with very small shear lips. The equating of the plane stress surface zone with a shear lip usually found in the metals literature appears to be unnecessary in the case of polymers, and indeed, shear lips are totally absent in polystyrene. It seems most likely that plane stress may be associated with bulk yielding while the constrained plane strain state results in localized yielding (shear bands) or the formation of a craze.

The extent of the plane stress zone from the surface is taken as the plastic zone size:

$$r_{y2} = \frac{1}{2\pi} \frac{K_{C2}^2}{\sigma_y^2}$$
(4)

and when a specimen of thickness H fractures, the apparent toughness K_C is made up of K_{C2} in the surface zones of width r_{y2} with K_{C1} acting in the central section.

i.e.
$$HK_{C} = 2r_{y2}K_{C2} + (H - 2r_{y2})K_{C1}$$

and $K_{C} = K_{C1} + \frac{2r_{y2}}{H}(K_{C2} - K_{C1}).$ (5)

Clearly, $K_{\mathbf{C}}$ decreases with increasing H tending to K_{C1} for large thicknesses and for $H \leq 2r_{y2}$, $K_{C} =$ K_{C2} . In fact, this condition is not usually reached in polymers undergoing the normal yielding process, since for $H < 4r_{v2}$ the central plane strain region is no longer sustained and ductile fracture occurs with substantial thickness reductions [6]. For toughened polystyrenes, however, the crazing process takes place without any appreciable lateral contraction (Poisson's ratio $\simeq 0$) and so it is possible to achieve $K_{C} = K_{C2}$ without throughthickness contractions. Although this problem may be avoided, the further one of plastic zone size is difficult with these materials. For the expressions for deriving $K_{\rm C}$ to be valid, $r_{\rm v2}$ must be less than the crack length and must also not be close enough to the edge of the specimen to cause the onset of gross yielding. Observed plastic zone sizes are typically around 10 mm in small specimens so that for 20 mm cracks in SEN tests the plastic zone is quite near the back edge in a 50 mm sample. This led to the problems of gross yielding mentioned earlier [7]. The use of large specimens seems to overcome this problem but the true zone sizes are about 3 mm indicating that the observed 10 mm sizes are probably augmented by net section stress effects. To obtain reliable values it seems easiest to test large thicknesses (e.g. > 10mm) but these are difficult to produce. The effective thickness concept of the surface notch overcomes this successfully and is the reason for its inclusion here.

3. Data on polystyrene

A series of SEN tests were performed on 3 mm thick polystyrene with sharp notches induced by fatigue and some by the use of environments. Both methods produced sharp notches and gave identical data [4] which are shown in Fig. 2. A check on thickness effects was made at 20° C by testing some SN specimens with $H' \simeq 13$ mm. The data are shown in Fig. 3 indicating excellent consistency and a value in agreement with the SEN result. The data in Fig. 2 are almost constant at about $K_{\rm C} = 1$ MN m^{-3/2} with a slight increase over the range 20 to -40° C. There is a small β peak in this



Figure 2 Effect of temperature on the fracture toughness of polystyrene for $\dot{x} = 0.05$ cm min⁻¹.



Figure 3 Results of surface notched polystyrene at $\dot{x} = 0.05 \text{ cm min}^{-1}$.

region [10] which gives rise to this effect. The data for PMMA [5] are also shown for comparison indicating a stronger effect from a more pronounced β peak.

4. Data on rubber modified polystyrene

A toughened polystyrene with a rubber content of about 4%* by weight was used in 6 mm thick sheets. SEN tests on 50 mm wide specimens gave ductile failure at 20° C [7] but specimens at 150 mm did indicate a value of $K_{\rm C} = 2.6 \,\rm MN \,m^{-3/2}$ at a cross-head rate $\dot{x} = 0.5 \text{ cm min}^{-1}$ [9]. Tests were performed here on SN specimens with $H' \simeq$ 13 mm and brittle fractures were obtained for crack depths deeper than 2.5 mm (for d < 2.5 mm gross yielding occurred). The data for three \dot{x} values are shown in Fig. 4 indicating a much enchanced rate effect as compared with polystyrene. The specimens showed much less whitening around the fracture than SEN specimens as would be expected from the lower $K_{\rm C}$ values. The presence of slow crack growth was difficult to establish from the fracture surface because of the presence of the crazed zone which was apparent on the surface. The $K_{\mathbf{C}}$ values are computed from the original crack dimensions and are probably slightly low but represent the onset of crack growth rather than a true instability value.

The experiments were then repeated at temperatures down to -120° C at $\dot{x} = 0.5$ cm min⁻¹ for both SEN and SN specimens and the resulting



Figure 4 Results for SN rubber modifed polystyrene at 20° C.

^{*} Kindly supplied by BP Chemicals (UK) Limited.



Figure 5 Fracture toughness for rubber modified polystyrene at x = 0.5 cm min⁻¹.

 $K_{\rm C}$ values as a function of temperature are shown in Fig. 5. The SN specimens gave brittle fracture over the whole temperature range while SEN only gave reliable data for -60° C and below. The fractures at higher temperatures showed considerable evidence of ductility with the $K_{\rm C}$ representation being increasingly unsatisfactory as the temperature increased above -60° C. The large specimen result at 20° C is also shown and the broken line linking this with the -60° C data gives a curve of similar form to the SN which is believed to represent the true brittle behaviour. Yield stress data were determined in simple tension over the same temperature range at a strain-rate of $5 \times 10^{=3}$ sec⁻¹ and these are shown in Fig. 6.

The $K_{\rm C}$ data show the expected pronounced thickness effect and by using Equation 4 and 5 together with $\sigma_{\rm y}$ it is possible to deduce $K_{\rm C2}$ and $K_{\rm C1}$. It is assumed here, however, that for full



Figure 6 Effect of temperature on tensile yield stress of rubber modified polystyrene.



Figure 7 Effect of temperature on K_{C_2} and K_{C_1} of rubber modified polystyrene at $\dot{x} = 0.5$ cm min⁻¹.

constraint the toughening mechanism will be inactive and so K_{C1} will be approximately that of the polystyrene matrix. Thus, if K_{C1} is taken as the polystyrene values from Fig. 2, then it is possible to deduce K_{C2} values for both SN and SEN data which may be compared. The results of this computation are shown in Fig. 7 and there is clearly close agreement of the two K_{C2} values supporting the K_{C1} assumption. The K_{C2} data increase steadily with temperature down to about -80° C after which there is a decrease. This form is consistent with polycarbonate [6] and in this case reflects the relaxation peak of the rubber phase [2].

That the K_{C2} data are a reflection of σ_y is shown in Fig. 8 where K_{C2} is plotted versus σ_y and there is proportionality with $K_{C2} \rightarrow K_{C1}$ as $\sigma_y \rightarrow 0$. The data at 20° C are also plotted in which σ_y varies with rate and this is consistent. Below -80° C the relaxation of the rubber becomes less active and the interaction of K_{C2} and σ_y decreases since it depends on the relaxation process. A similar effect was observed in polycarbonate above -40° C. Polycarbonate [6] and PMMA [5] also showed the general proportionality of K_C and σ_y within a relaxation region. The plastic zone size r_{y2} decreases from about 2.5 mm at 20° C to around 2 mm at low temperatures as shown in Fig. 8. For the SEN tests using 6 mm thick sheet, K_C



Figure 8 K_{C_2} and r_{y_2} as a function of σ_y .

would be expected to be very close to K_{C2} and this was the case for the wide plate test at 20° C.

For both PMMA and polycarbonate the yield strain was approximately constant with temperature and it was found that the crack opening displacement:

$$u = \frac{K_{\rm C2}^2}{\sigma_{\rm v}E}$$

was constant also (1.6 and $37 \,\mu\text{m}$, respectively). Table I shows modulus, yield stress, yield strain $(e_y = \sigma_y/E)$ (at a strain-rate of $5 \times 10^{-3} \,\text{sec}^{-1}$) and computed *u* values for the data obtained here. e_y increases with decreasing temperature down to about -60° C and then remains constant and *u* follows this trend. This is consistent with the general conclusion that *u* reflects the state of strain at the crack tip. It should be noted that an

TABLE I

Temperature (°C)	<i>E</i> (GN m ⁻²)	σ_{ys} (MN m ⁻²)	e _y	u(µm)
20	3.9	20.5	0.0052	84
0	4.1	27.8	0.0067	107
- 20	4.3	35.2	0.0082	128
- 40	4.6	46.2	0.0100	143
- 60	4.9	53.5	0.0110	152
- 80	5.4	62.0	0.0114	150
-100	5.8	66.9	0.0114	132

arbitrary choice of strain-rate is made here since the crack speed is not known and it would be required for an exact determination [5]. The absolute value of u may, therefore, be incorrect but the trend is likely to be unaltered.

5. Conclusion

The use of the SN test together with SEN has given true brittle fractures for rubber modified polystyrene over the whole temperature range -120 to 20° C. The difference in the two sets of data further supports the idea of plane stress and plane strain K_{C} values. It appears from the data that K_{C1} is the value for the matrix material and that constraint inhibits the crazing process induced by the rubber phase. This is predicted by the crazing criterion proposed by Oxborough and Bowden [11] for which the stress necessary to produce crazing becomes very large as Poisson's ratio tends to 0.5 under constraint. The toughening takes place in the plane stress region and the relaxation processes inherent in the rubber are reflected in K_{C2} . The yield strain varies over part of the temperature range and this is reflected in a variation in crack opening displacement.

The parallel with the data on polycarbonate is instructive in describing the role of the rubber phase. The small rubber content introduces a relaxation process into the polystyrene which is active in the low temperature region and models the β process in polycarbonate. The process is only effective under plane stress conditions, however, again as in polycarbonate. The generation of crazes by the rubber particles is reflected in the decrease of the yield stress as compared to polystyrene and the consequent increase in the plane stress region. This is consistent with the generally accepted view of the process [1] and gives a basis for a quantitative assessment of toughening.

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