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A fracture mechanics analysis of the effect of rubber content on rubber-modified polystyrene

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Consideration is given to achieving test conditons in which linear elastic fracture mechanics may be employed to measure toughness in these rather ductile materials. This is achieved by testing wide specimens (100 mm) and these are employed to measure K_c over a wide range of temperatures for materials with several rubber levels. It is shown that both K_c and the yield stress are determined by the extent of crazing and that, at the low rates used here, the full crazing capacity of the polystyrene is utilized by the addition of about 1% of rubber.

1. Introduction

Rubber-modified polymers are designed to provide materials of substantial toughness while retaining an acceptable degree of stiffness [1]. Blends of polystyrene and rubber have been quite successful in this regard, and there is a good deal of interest in defining this toughness accurately and establishing how certain geometric and processing parameters affect its value. The use of linear elastic fracture mechanics (LEFM) has received some attention [2–4] and it was soon noted that these materials presented a basic problem in that the toughness was enhanced by reducing the yield stress, thus increasing the extent of plastic flow and consequently violating the conditions for LEFM to apply [3, 5]. There have been several attempts to apply non-linear fracture theory to deal with this, including crack opening displacement and J_c [3, 5]. These have been reasonably successful, but it is considered to be preferable, especially for comparison purposes, to use a conventional fracture toughness K_c (or G_c) if this is possible. This paper sets out to define some test conditions which achieve this and then uses these to investigate the important practical parameter of rubber content.

2. Analysis of test conditions

Rubber-modified polystyrenes have rather special deformation properties in that the enhancement of toughness is achieved by the introduction of many crazes around the rubber particles [1]. These crazes form by the creation of voids so that there is a substantial apparent volume increase in the material as it deforms. For the case of a single-edge notch (SEN) tension specimen, there is considerable whitening around the crack tip but almost no lateral contraction, so that substantial crazed zones may be formed while a crack may grow through them in essentially a flat, plane strain mode. In general, one must consider the comparison of the zone size, r_p, to both the specimen thickness, B, width, D, and crack length, a, to determine if valid LEFM conditions pertain.

The thickness condition is generally that $B > 4r_p$ for gross yielding within the zone to be avoided, but for these materials this is not applicable and flat fractures do occur down to very small B values. There are thickness effects, however, since plane strain conditions suppress craze formation and tend to reduce the toughness to the basic matrix value [4]. This effect can be modelled by supposing that there is a toughness associated with plane stress conditions, K_{c2}, which incorporates the full effects of crazing, while the plane strain state gives a value of K_{c1} which is the basic matrix value. A specimen of thickness B will, therefore, have an average value K_c' made up from a sandwich with a centre value of K_{c1} and two surface layers of K_{c2} and a thickness given by:

$$r_{p2} = \frac{1}{2\pi}\frac{K_{c2}^2}{\sigma_y^2} \tag{1}$$

where σ_y is the yield stress.

We may write this average as:

$$K_c' \doteq K_{c1} + \frac{2r_{p2}}{B}(K_{c2} - K_{c1}). \tag{2}$$

This relationship has been shown to be useful in describing rubber-modified polymers [4] and will be employed later here. It should be noted that for large B values, $K_c' \to K_{c1}$, and that for $B < 2r_{p2}$, $K_c' = K_{c2}$, the completely crazed value.

Specimen width effects have received little attention using these materials. The basic LEFM relationship is:

$$K_c = Y\sigma\sqrt{a} \tag{3}$$

where σ is the gross stress, and:

$$Y = 1.99 - 0.41\,x + 18.7\,x^2 - 38.48\,x^3 + 53.85\,x^4$$

(for the SEN specimen) with $x = a/D$. Y describes the enhancement of stress in the net section, both from the free edge and the finite width, D, but it assumes that all the deformations are elastic. For $x < 0.5$ and $\sigma/\sigma_y < 0.75$, this works well and, for small amounts of plasticity, may be extended by adding on a correction factor to a of $r_p/2$. When small crack lengths are used $\sigma \to \sigma_y$, and the limiting condition is given from Equation 3 when:

$$\bar{a} = \frac{1}{Y^2}\frac{K_c^2}{\sigma_y^2}.$$

Now, the plastic zone size is given by:

$$r_p = \frac{1}{2\pi}\frac{K_c^2}{\sigma_y^2}$$

and since $a/D \ll 1$ here, we may write $Y^2 \simeq \pi$ for compatability with the relationship for r_p, so that:

$$\bar{a} = 2r_p. \tag{4}$$

For $a < \bar{a}$, there will be gross yielding in the specimen. A more demanding condition is also possible since yielding may occur in the ligament of the specimen and, in this case:

$$\sigma_y = \frac{\sigma}{1 - \hat{a}/D}.$$

Although x may not be small in this case, Y^2 will not be used since it is not accurate for the substantial degrees of plasticity modelled here, and so we shall retain the assumption of $Y^2 = \pi$ and, by utilizing Equation 3 again, we have a condition for net section yield given by:

$$\frac{\hat{a}}{D}\left(1 - \frac{\hat{a}}{D}\right)^2 = \frac{2r_p}{D} = \frac{\bar{a}}{D}. \tag{5}$$

Fig. 1 shows this relationship and the gross yield condition (the broken line) for $\hat{a} = \bar{a}$*. For $\bar{a}/D < 4/27$, there are two values of $\hat{a}/D$: the first equivalent to gross yielding, the second to net section yielding. For $\bar{a}/D > 4/27$, there are no solutions indicating that $\sigma/\sigma_y > 1$ for all crack lengths and it is impossible to achieve fracture at a below yield stress. A limiting D may, therefore, be defined below which only ductile failure is possible:

$$D_{\min} = \frac{27}{4}\bar{a} = \frac{54}{4}r_p = \frac{54}{8\pi}\frac{K_c^2}{\sigma_y^2}. \tag{6}$$

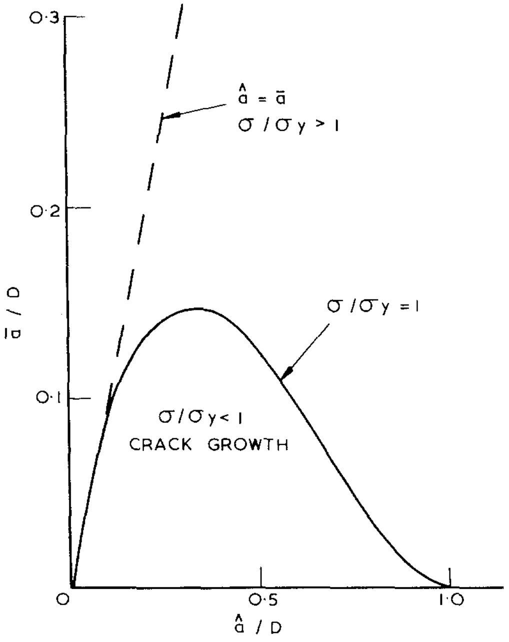

Figure 1 Yielding condition in the notched tension test.

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* A similar relationship has been used previously, e.g. [6].

It should be noted that for $\bar{a}/D < 0.1$ there is a considerable region of crack growth before net section yielding occurs and causes arrest. For $\bar{a}/D > 0.1$, this growth is very limited in extent. For brittle materials, $\bar{a}/D << 1$ and so no effect is observed but for the tougher materials, as described here, the effect could be significant.

3. Materials and test procedure

Two types of rubber-modified polystyrene material were used. Firstly, the effect of the specimen geometry was investigated, using a commercial grade of high impact polystyrene, which was supplied in sheets of 6.4 mm nominal thickness. The second part of the work was devoted to the study of the effect of temperature and rubber content on the fracture toughness and yield stress of a series of specially prepared grades which were manufactured in sheets of 3.2 mm nominal thickness. These materials were produced by back-blending a 5.55% rubber-modified grade with the parent crystal polystyrene matrix to give nominal rubber contents of 0, 1.11, 2.22, 3.33 and 5.55% rubber. The back-blending process was carefully controlled so as to minimize any variation in the properties of the composite components. The materials obtained were, therefore, reasonably representative of composites having identical component properties, but varying in the quantity of the rubber incorporated.

Single-edge notched (SEN) specimens were machined to the required dimensions and sharp notches of various lengths were then cut in one edge with the aid of a single point fly cutter of a wedge angle of about 3° and a tip radius of less than 0.002 mm. A cross-head speed of $0.05\,\text{cm min}^{-1}$ was selected for fracture toughness tests to allow sufficient time for craze formation and crack initiation to be carefully observed and recorded (the toughness is rate-dependent for these materials [3, 4]). Experiments on both notched and dumbbell specimens for yield stress determination were carried out in a temperature cabinet which was fitted to an Instron testing machine. The cabinet contained electrical heating elements to maintain temperatures greater than ambient, and liquid nitrogen was vaporized and circulated inside the chamber for low temperature experiments. Temperature control was achieved with an accuracy of about ± 1° C over the range of – 140 to + 60° C, utilizing a Eurotherm control unit.

4. Geometry effects

The particular grade of material was chosen for these tests because it had a rather low yield stress (nominally $15.0\,\text{MN m}^{-3/2}$) and was known to give substantial ductility effects. Tests were performed at 20° C on two batches of specimens, one with $D = 50$ mm and the other with $D = 100$ mm. The data obtained is shown in Fig. 2 plotted as σ versus a/D and the 50 mm results clearly fall on the fully ductile line using $\sigma_y = 12.5\,\text{MN m}^{-2}$. For the 100 mm specimens, there is a considerable reduction in stress and the points follow a line for $K_c = 2.50\,\text{MN m}^{-3/2}$ very closely. Assuming now that this is the correct value of K_c, then $r_p = 6.4$ mm and D_{min} is 86 mm from Equation 6. Thus, the 50 mm specimens would be expected to be completely ductile, as they are, while the 100 mm should show crack growth. The transition to ductile fracture in this case is at $a/D = 0.18$ from Fig. 1, which is in close agreement with the result shown in Fig. 2.

A series of tests were also performed with 100 mm wide specimens coated on both sides with a layer of a brittle epoxy resin of 0.25 mm thick. It was known that the presence of such layers can render some materials brittle [7]. In this case, it was found that the load–deflection diagram was less curved, indicating a suppression of craze formation, but that failure occurred at identical loads to the uncoated case such that the value of K_c remained the same. The fracture appeared to initiate within the main material but, in this case, there was extremely rapid crack propagation as opposed to rather slow growth for uncoated specimens. The load decreased almost to zero and the crack arrested to be followed by the load increasing again to some lower value to give further rapid growth and so to total failure by several of these steps.

A possible explanation of this effect is that the layers provide an additional constraint which suppresses craze formation and hence increases the effective yield stress. The toughness will tend to increase, but only slightly because, although K_{c2} increases, r_{p2} will decrease (see Equations 1 and 2). The decrease in r_p is the key factor, however, since this will decrease the $2r_p/D$ ratio and thus greatly increasing the range of crack growth condition crack lengths (see Fig. 1). Thus, the uncoated material does give crack growth, but only for a short range of crack lengths, while a

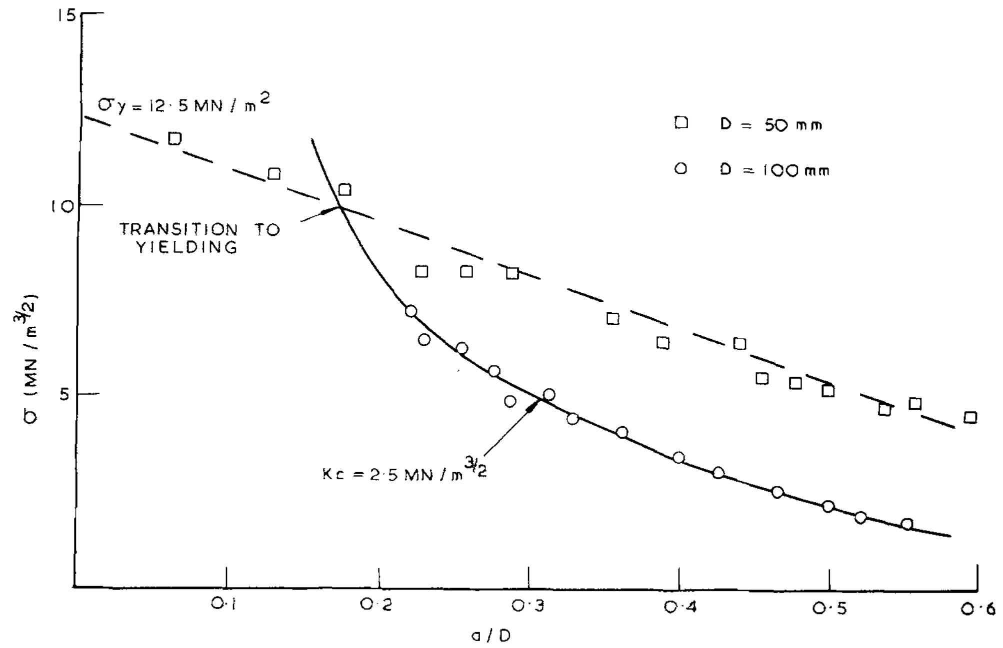

Figure 2 SEN data for specimen widths at 50 and 100 mm.

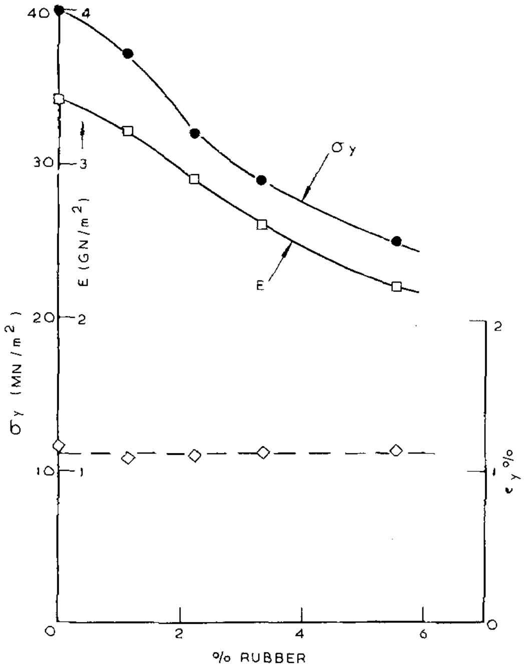

Figure 3 Modulus, yield stress and yield strain as a function of rubber content at 20° C.

modest increase in σ_y can greatly increase this, thus making the fracture more obviously brittle.

5. Rubber content and temperature

The series of specially prepared materials with different rubber contents were first tested in the form of moulded bars to determine the yield stress at a range of temperatures. For zero rubber content, i.e. the basic polystyrene, the maximum load value corresponds to the onset of substantial crazing which is soon followed by a brittle fracture. For the modified materials, there is a clear maximum in the load–deflection curve after crazing starts followed by considerable elongation (up to 50%) before a ductile rupture. At 20° C, both the yield stress and modulus were measured and these are shown plotted against rubber content in Fig. 3. E and σ_y vary in the same manner and this is borne out by the consistency of the yield strain (σ_y/E) at 1.1%. Since the stiffness of the rubber is low compared with the polystyrene, a percentage modulus drop of around that of the rubber content would be expected from a simple law of mixtures. In fact, the decrease is much greater, being 37.5% for 5.5% rubber. This indicates an effective rubber content of 37.5% which is consistent with the concept of polystyrene particles included in the rubber [1], thereby increasing their effective size. The implication here is that 32% of the polystyrene is included in the rubber.

The yield stress as a function of temperature for the various rubber contents is shown in Fig. 4. The polystyrene shows a tendency for a more rapid change with temperature in the range +30 to −30° C, and this is reflected in the rubber-modified materials. The relative effect of the rub-

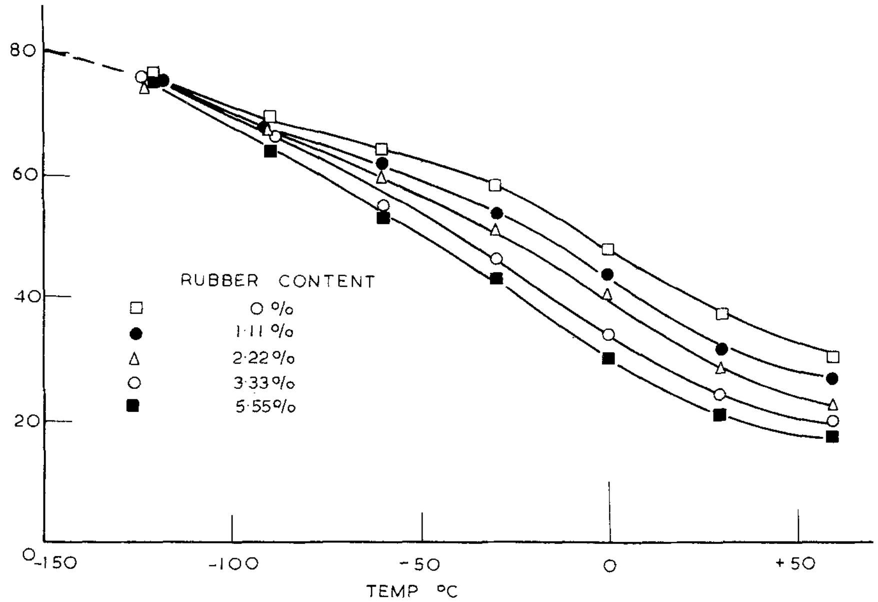

Figure 4 Yield stress for various rubber contents as a function of temperature.

ber decreases with decreasing temperature, falling to virtually zero at −120° C.

A mechancial loss analysis [8] of these materials revealed a very small peak in the polystyrene at −40° C of 0.003 which may reflect a process within the polymer [9] or, more likely, be due to the addition of mineral oil as a processing aid. It is believed that this is the effect which is seen in the yield data. For the rubber-modified materials, a peak from the rubber occurs at −80° C and increases in proportion to the rubber content, giving 0.015 for 5.5% rubber. These rubber peaks do not appear to be reflected in the yield stress data, but below −80° C, which is the glass transition temperature of the rubber, the particles cease to be effective craze generators.

Fracture tests were performed over the temperature range −120 to +60° C and Fig. 5 shows some of the results. The data for the polystyrene increases with decreasing temperature with a definite peak around −40° C. One set of data is shown for a modified material (3.33% rubber) and the value of K_c for both crack initiation and final maximum load at rupture is shown. The

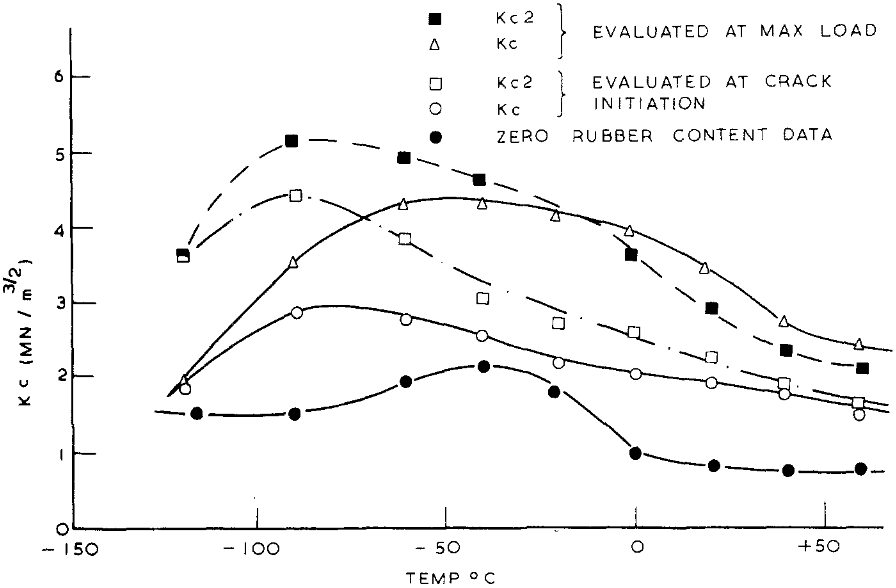

Figure 5 Toughness as a function of temperature for 3.33% and 0 rubber content.

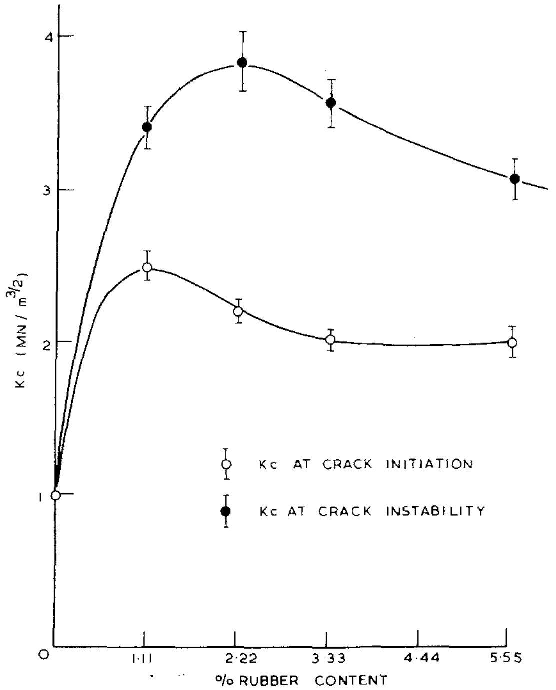

Figure 6 Variation of fracture toughness with rubber content at 20° C.

initiation values are substantially increased above the basic polystyrene values although around -40° C, the difference is rather small. Also shown in Fig. 5 are the K_{c2} values at both crack initiation and final rupture as computed from Equations 1 and 2. At initiation, $K_{c2} > K_c'$, so that there is always some material in the plane strain state. At final load, this only occurs below -40° C and at higher temperatures, the failure is at K_{c2}.

Fig. 6 shows both initiation and instability K_c values as a function of rubber content at 20° C and it is evident that the toughness increases very rapidly with rubber content and then declines thereafter. The mechanism implied by this is that the presence of rubber induces crazes and that, in these rather slow tests, the crazing capacity of the material is utilized fully by a very small rubber content, probably around 1%. The decline in K_c thereafter suggests a dependence similar to the yield stress (see Fig. 3).

Fig. 7 shows all the K_{c2} values, together with the polystyrene data, plotted against the appropriate yield stress. For the modified materials, it is clear that the toughness is determined by the yield stress and that this dependency is not influenced by the actual rubber content, providing it is greater than 1%. This confirms the idea of fully developed crazing which is reflected in both K_c and σ_y. The peak in the polystyrene K_{c1} data is again apparent and here it can be seen reflected in the K_{c2} data also. A possible explanation of this phenomenon is that, around the freezing temperature of the added oil (-40° C), the oil globules have sufficient stiffness to form crazes, as do rubber particles. Thus, over a fairly narrow temperature (and therefore σ_y) band, there is an increase in craze generation. The mechanism is quite distinct from that of the rubber particles since it can be seen to be additive even in the craze saturation condition. Visco-elastic losses are unlikely to be very large for a process with such a low loss peak and it may be that a very fine dispersion of oil generates an array of much finer crazes than the rubber particles. As noted previously, there is a quite distinct change in slope in the σ_y versus temperature data (Fig. 4) at about -40° C which would be consistent with this, since the additional crazing would depress the yield stress at higher temperatures.

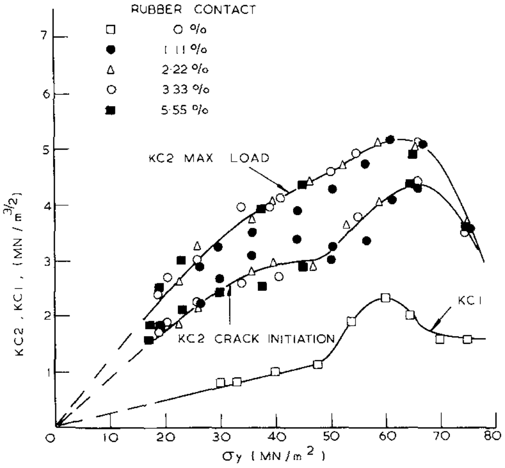

Figure 7 Plane strain (K_{c1}) and plain stress (K_{c2}) fracture toughness values as a function of yield stress.

The effect of the rubber induced crazes can be illustrated by subtracting K_{c1} from K_{c2} and Fig. 8 shows points obtained by using the average lines drawn in Fig. 7. There is some scatter but they do suggest linearity up to about $\sigma_y = 30\,\mathrm{MN\,m^{-2}}$ with the addition remaining constant for higher σ_y values. The toughness increase is brought about by an increase in elongation to fracture with the addition of rubber which more than compensates for the decrease in σ_y. Fig. 9 shows tensile elongation ϵ data obtained at 20° C as a function of rubber content which can be seen to increase linearly. The energy absorbed would be proportional to $\epsilon\sigma_y$ but expressed in terms of K_c

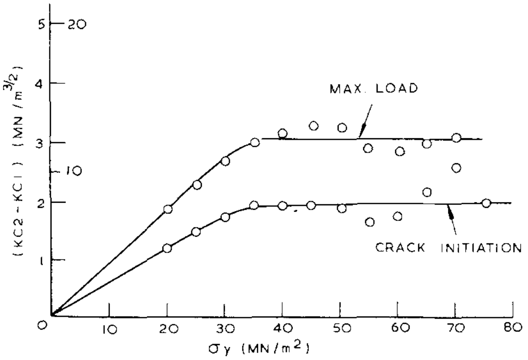

Figure 8 $(K_{c_2} - K_{c_1})$ as a function of yield stress.

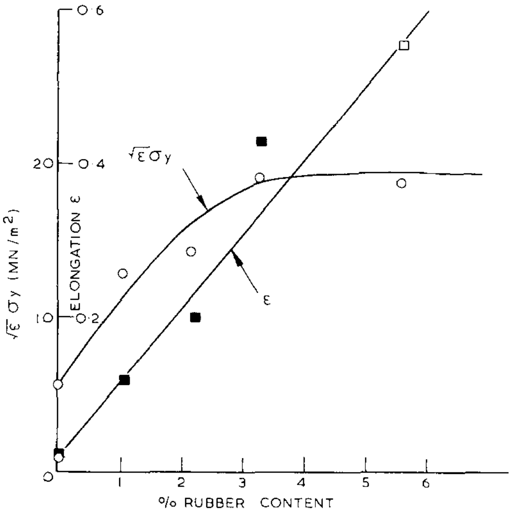

Figure 9 Elongation ϵ and $\sqrt{\epsilon}$, σ_y as a function of rubber content.

it would be proportional to $\sqrt{\epsilon\sigma_y}$ (assuming ϵ_y is constant) and this is also shown in Fig. 9. Again, there is a levelling out suggesting a saturation effect.

The plastic zone size, r_{p2}, never exceeds 2.5 mm here, so that the specimen width of 100 mm was more than adequate to ensure the applicability of LEFM, since the critical width (D_{min}) is 34 mm.

6. Conclusions

The results on the low yield stress material have illustrated that it is possible to devise a test geometry which will give valid K_c values so that conventional LEFM analysis may be used. The use of this method on the series of specially prepared materials shows that the toughness and the yield stress are governed by the same craze formation mechanisms and that, in this case, there is one associated with the basic polystyrene, probably due to added mineral oil, and a second due to the rubber. In the slow tests used here, the crazing capacity of the material is utilized by the addition of about 1% rubber and that for rubber contents which are greater than this, there is a decline in toughness. This is in contrast to impact test behaviour [8, 10] where there is a linear increase of toughness with rubber content since only part of the crazing capacity for each particle can be utilized in the testing time, and so the larger the number of particles, the greater the crazing. The choice of rubber content for a practical application must, therefore, be a compromise between long-term toughness requiring very little rubber and impact conditions where much more is needed.

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