JOURNAL OF MATERIALS SCIENCE 12 (1977) 1376–1382

The effect of temperature on the fracture of two partially crystalline polymers; polypropylene and nylon

Y. W. MAI, J. G. WILLIAMS
Department of Mechanical Engineering, Imperial College of Science and Technology, London, UK

Brittle fractures in polypropylene and nylon were obtained in the temperature range -180 to $-10°$ C and -160 to $20°$ C, respectively. The fracture toughness results of surface notches and single edge notches showed a remarkable thickness dependence. This effect was explained in terms of a plane strain (K_{C1}) and a plane stress (K_{C2}) toughness value using a bi-modal fracture analysis. While K_{C1} in both materials was generally insensitive to temperature, K_{C2} was temperature dependent showing substantial visco-elastic effects. In polypropylene, the linear relationship between K_{C2} and temperature (T) was associated with the β and γ processes over the same temperature range. In nylon, there was a one-to-one correspondence between the K_{C2} changes and the tan δ peaks due to the β and γ relaxation processes. Using a modified crack tip opening displacement (u) equation, i.e. $u = ((K_{C2} - K_{C1})/\sigma_y)^2 e_y$, where σ_y and e_y are the yield stress and yield strain respectively, a constant u criterion was found to describe the fracture behaviour of both materials in the temperature ranges of the β and γ processes.

1. Introduction

The effect of temperature on the fracture of polymeric materials has been extensively studied recently [1–4]. These include brittle materials like PMMA [1–4] and polystyrene [5], as well as relatively ductile and tough polymers like polycarbonate [1] and rubber modified polystyrene [3]. In the experimental investigations [1–3] using both single edge notched (SEN) and surface notched (SN) tension specimens, and over the temperature range 20 to $-120°$ C, it was found that the brittle fracture data for PMMA and polystyrene did not display any thickness effect. However, such an effect was evident for rubber-modified polystyrene and polycarbonate. A bi-modal fracture analysis in terms of a plane stress fracture toughness (K_{C2}) and a plane strain value (K_{C1}) was shown to give an adequate explanation of the observed experimental results [2, 3].

In the study reported here, fracture experiments were performed on polypropylene and nylon over the temperature range -180 to $20°$ C. Brittle fracture was achieved through the use of surface notched specimens. These data, together with those derived from SEN test pieces and coupled with the temperature dependent yield stress (σ_y), have given values of K_{C2} and K_{C1}.

2. Bi-modal fracture analysis

When brittle fracture occurs in a cracked specimen under uniform tension σ, the apparent fracture toughness (K_C) is given by

$$K_C = \sigma\sqrt{a}Y, \tag{1}$$

where a is the characteristics flaw size and Y the geometric correction factor depending on specimen geometry. For SEN specimens, Y can be found from the boundary collocation results of Brown and Srawley [6], and for SN testpieces with semi-elliptic part-through cracks

$$Y^2 = \frac{1.21\pi}{\phi^2 - 0.212\,(\sigma/\sigma_y)^2} \tag{2a}$$

and

$$\phi = \int_0^{\pi/2} \left[1 - \left(\frac{b^2 - a^2}{a^2}\right) \sin^2\theta\right]^{1/2} d\theta \quad (2b)$$

In Equation 2, σ_y is the tensile yield stress and b the semi-major axis of the semi-elliptic cracks.

In tough polymers such as those studied in this work, it was found useful in subsequent analysis to partition the brittle fracture toughness (K_C) into two toughness sources, one the plane strain (K_{C1}) and the other plane stress (K_{C2}). By assuming the plane stress toughness contribution to be effective over the skins of typical dimension $2r_{y2}$, and plane strain to prevail over the remaining middle portion ($H - 2r_{y2}$) of the total thickness H of the test specimen, we have

$$HK_C = (H - 2r_{y2})K_{C1} + 2r_{y2}K_{C2} \quad (3a)$$

$$r_{y2} = \frac{1}{2\pi}\left(\frac{K_{C2}}{\sigma_y}\right)^2. \quad (3b)$$

Thus

$$K_C = K_{C1} + \frac{K_{C2}^2(K_{C2} - K_{C1})}{\pi\sigma_y^2 H}. \quad (3c)$$

When H is sufficiently large, $K_C \to K_{C1}$ and in the absence of sufficient thickness, surface notched specimens may be employed to yield apparent thicknesses H' which are larger than their physical thicknesses. H' may be obtained by equating the area of the circular arc surface crack to that of a thorough crack having a uniform crack depth a. Thus

$$H' = \frac{R^2}{a}\left[\sin^{-1}\left(\frac{b}{R}\right) - \frac{b}{R}(1 - a/R\right], \quad (4)$$

where R is the radius of curvature and is related to b and a by

$$b^2 = a(2R - a). \quad (5)$$

For $b/R \ll 1$, Equations 4 and 5 yield:

$$H' \simeq \tfrac{4}{3}b. \quad (6)$$

In this respect, it is obvious that surface notched tension specimens are best used for investigating thickness effects on fracture.

Irwin [7] suggested that the ductile to brittle transition occurs when the plastic zone size ($2r_{y2}$) is smaller than half the specimen thickness, so that

$$H > 4r_{y2}. \quad (7)$$

When $H < 4r_{y2}$, ductile fracture predominates since there is insufficient material under constraint to give the low K_{C1} contribution to toughness.

Provided there is no pronounced thickness dependence of σ_y, any two sets of brittle fracture data (K_C' and K_C'') from SEN or SN specimens corresponding to two different thicknesses H_1 and H_2, may be used with Equation 3c to give appropriate expressions for K_{C1} and K_{C2}. Thus

$$K_{C1} = \frac{H_1K_C' - H_2K_C''}{H_1 - H_2} \quad (8)$$

and

$$K_{C2}^3 - K_{C2}^2K_{C1} - \pi\sigma_y^2H_1(K_C' - K_{C1}) = 0. \quad (9)$$

3. Test procedure

Rectangular specimens of 6 mm thick polypropylene (ICI, compression moulded) 150 mm by 50 mm, and of 5 mm thick nylon (ICI Maranyl AD 151, type 66, injection moulded) 235 mm by 86 mm, were used for both single edge and surface notches. The starter cracks were introduced to the specimens by machining with a dead sharp fly cutter having tip radius less than 0.5 μm. The cutter radius for the surface notches was varied between 15 and 50 mm to give a wide range of apparent thicknesses. To obtain an almost straight and sharp crack tip for the single edge notches, a large cutter radius was used. The nylon specimens were all conditioned to a water content of 0.2% before the notching was done.

Fracture experiments were then conducted on these SEN and SN specimens in an Instron testing machine equipped with a temperature controlled box. The temperature within the range 20 to

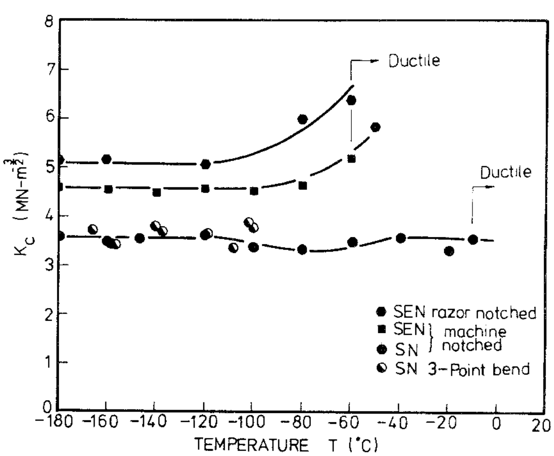

Figure 1 **Fracture toughness for SN and SEN polypropylene.**

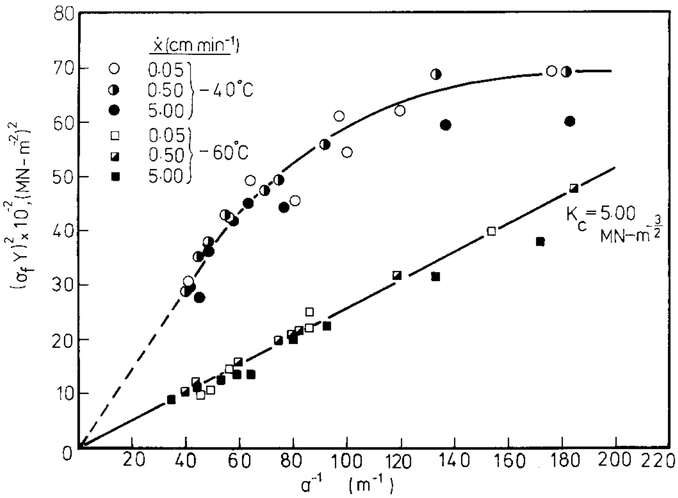

Figure 2 Results for SEN polypropylene at −40 and −60° C.

−180° C was controlled to an accuracy of ±1° C by a "Eurotherm" control unit and the actual temperature of the specimen measured by a thermocouple adhered near the crack tip region.

4. Results

4.1. Results on polypropylene

At a cross-head of $5\,\text{mm min}^{-1}$, SN specimens displayed brittle fracture in the temperature range −40 to −180° C and showed some ductility from −10 to −20° C and became ductile above −10° C. SEN specimens exhibited gross yielding at about −40° C and gave brittle fracture at −60° C and below. The variation of K_C (average of at least five specimens) with temperature is shown in Fig. 1. Some of the SN specimens were also used in three-point bending. Brittle fracture was obtained only below −60 and above −40° C all fractures were completely ductile. These K_C data are also superimposed in Fig. 1, which show good agreement with the SN tension results for temperatures below −80° C.

Unlike rubber-modified polystyrene [3], rate affects on the K_C values of polypropylene were insignificant. Fig. 2 shows the SEN fracture data for three cross-head rates $\dot{x}$ at −40 and −60° C. Gross yielding was predominant at −40° C and brittle fracture at −60° C gave $K_C \simeq 5.00\,\text{MN m}^{-3/2}$. Fig. 3 records the brittle fracture data of SN specimens at −40° C at four $\dot{x}$ values. Again, rate effects were negligible. However, when $\sigma/\sigma_y > 0.70$, some experimental data begin to fall off the line corresponding to $K_C = 3.85\,\text{MN m}^{-3/2}$ and show some rate dependence at yield. This is not unexpected since the yield stress σ_y increases with the cross-head rate.

4.2. Results on nylon

Both SN and SEN specimens exhibited brittle fracture in the temperature range 20 to −160° C. The variation of K_C (average of at least three specimens) with temperature is given in Fig. 4. The trend of these data is similar to that of polypropylene in that the SN specimens give fairly constant K_C magnitudes with temperature and the SEN testpieces show increasing K_C values with temperature. These tests were conducted at $\dot{x} = 5.00$ mm min^{-1}.

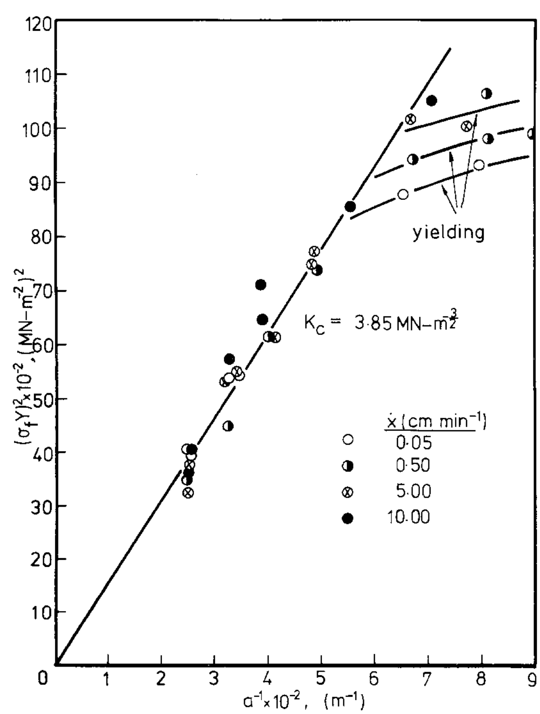

Figure 3 Results for SN polypropylene at −40° C.

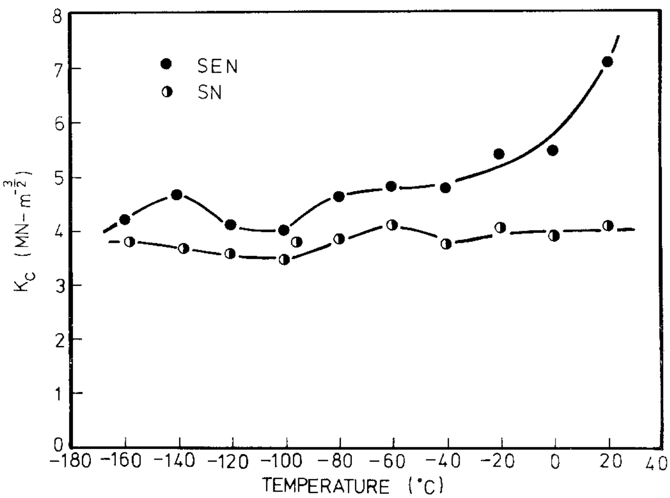

Figure 4 Fracture toughness for SEN and SN nylon (0.2% water content).

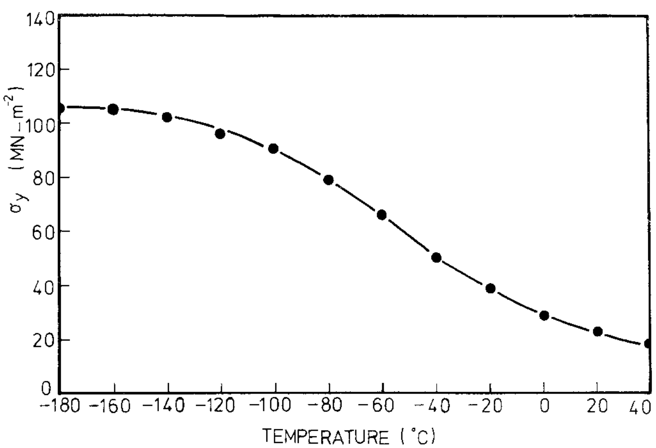

Figure 5 Yield stress as a function of temperature for polypropylene.

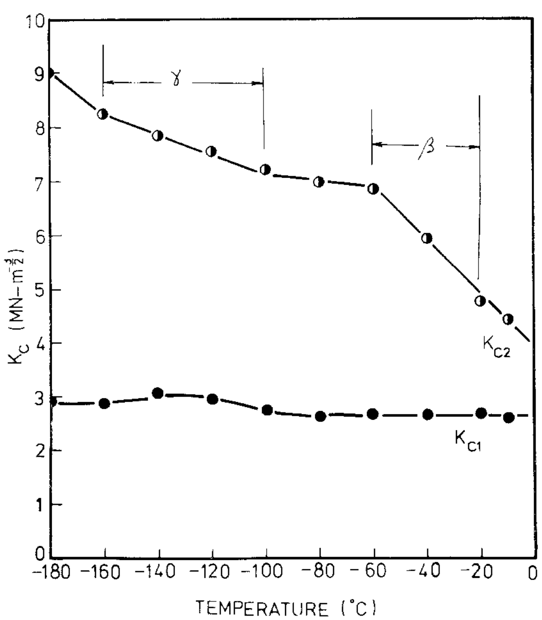

Figure 6 K_{C1} and K_{C2} as a function of temperature for polypropylene.

5. Discussion

5.1. Polypropylene

The lower K_C values obtained for SN specimens indicated the presence of a thickness effect on fracture. The apparent thickness H' of the 6 mm SN specimens was about 16 mm. Equations 8 and 9 together with σ_y, which is obtained separately and shown in Fig. 5, have enabled the determination of both K_{C1} and K_{C2} in the temperature range −180 to −10° C. These results are given in Fig. 6 and show that K_{C1} is approximately 2.70 MN m$^{-3/2}$ for all temperatures. The linear relationship between K_{C2} and T in the temperature range −10 to −60° C is a result of the pronounced β-relaxation process, the viscoelastic energy losses of which increase the K_{C2} values. Below −100° C, K_{C2} also increases linearly with temperature because of the viscoelastic effects of the somewhat less prominent γ-process. These results are similar to those observed in polycarbonate below −40° C [2].

TABLE I Variation of K_C and K_{C2} with H' at $-60°$ C for polypropylene

Apparent thickness H' (mm)	Fracture toughness		
	K_{C1} ($MN\,m^{-3/2}$) (assumed constant)	K_C ($MN\,m^{-3/2}$)	K_{C2} ($MN\,m^{-3/2}$)
5.26 (SEN)	2.73	6.08	6.95
13.29 (SN)	2.73	3.91	7.00
16.94 (SN)	2.73	3.64	6.94
19.89 (SN)	2.73	3.51	6.96
23.00 (SN)	2.73	3.38	6.92

Table I shows the effects of apparent thickness H' on K_C and K_{C2} at $-60°$ C when the yield stress of the material is $65\,MN\,m^{-2}$. While K_C increases with decreasing H' as expected, K_{C2} calculated from Equation 9 using $K_{C1} \doteq 2.73$ $MN\,m^{-3/2}$ seems to be independent of H'. These results therefore further supported the general validity of the effective thickness concept of surface notch specimens.

Previous work on PMMA [1], polycarbonate [2] and rubber-modified polystyrene [3] have shown that, in the temperature range of predominant viscoelastic relaxation processes, K_{C2} is linearly dependent on σ_y and that K_{C2} approaches K_{C1} when σ_y is close to zero. This was found also to be true for polypropylene as shown in Fig. 7 where the β- and γ-processes were clearly displayed. The small plateau found around the yield stress region of 65 and $88\,MN\,m^{-2}$ is a reflection of the constant K_{C2} values between -60 and $-100°$ C, where $\tan\delta$ is very small. Also shown in Fig. 7 is the variation of plastic zone size r_{y2} with σ_y (or indirectly with T). Thus, r_{y2} decreases from a magnitude of 3 mm at $-10°$ C to about 1 mm below $-100°$ C. The ductile/brittle transition on the SEN specimens occurred at $-60°$ C which gave H/r_{y2} approximately 3.5. This result is close to the limiting value of 4 as required by the criterion given in Equation 7.

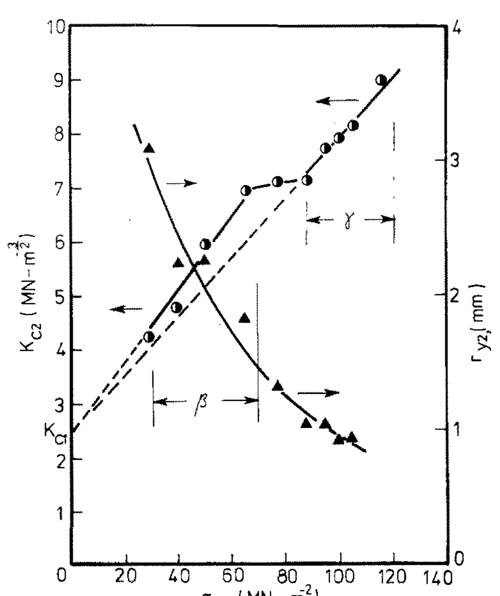

Figure 7 Variation of K_{C2} and r_{y2} with yield stress for polypropylene.

The yield strain e_y ($= \sigma_y/E$) increases slightly from 0.02 in the range -10 to $-60°$ C to 0.03 at $-100°$ C and below. When the crack tip opening displacement u is calculated from

$$u = \left(\frac{K_{C2}}{\sigma_y}\right)^2 e_y \tag{10}$$

the values varied with temperature. For example, within the β-range at $T = -60°$ C, $K_{C2} = 6.85$ $MN\,m^{-3/2}$, $\sigma_y = 66\,MN\,m^{-2}$ and $e_y = 0.02$, $u = 215\,\mu m$; and in the γ-range at $T = -120°$ C, $K_{C2} = 7.55\,MN\,m^{-3/2}$, $\sigma_y = 95\,MN\,m^{-2}$ and $e_y = 0.03$, $u = 188\,\mu m$. However, when u is obtained from the slopes, i.e. (u/e_y), of the two straight lines shown in Fig. 7, this gives a constant u value of $91\,\mu m$ for both the β- and γ-processes, although the yield strains are not the same in these temperature ranges. Since an invariant crack opening displacement is a useful fracture criterion which has been used previously to describe the fracture behaviour of PMMA [1] and polycarbonate [2], the present results seem to suggest that, if such a criterion is to be retained, u should be calculated from a modified form of Equation 10, i.e.

$$u = \left(\frac{K_{C2} - K_{C1}}{\sigma_y}\right)^2 e_y, \tag{11}$$

when describing bi-modal data. It seems likely that both K_{C2} and K_{C1} are proportional to σ_y but the critical displacement is much smaller for K_{C1} and is obscured by the larger variation in K_{C2}.

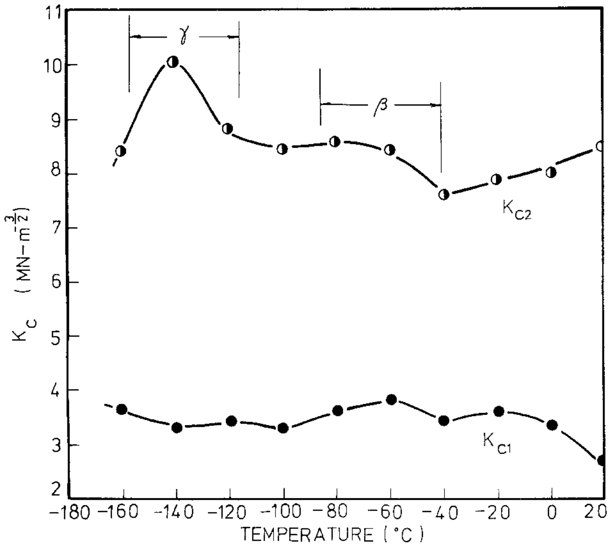

Figure 8 K_{C1} and K_{C2} as a function of temperature for nylon (0.2% water content).

5.2. Nylon

Like polypropylene, the lower K_C values given by the SN specimens having $H \simeq 18$ mm indicated the presence of a thickness effect. Despite some slight fluctuations, K_{C1} calculated from Equation 8 and shown in Fig. 8 is quite consistent in the temperature range 20 to $-160°$ C, giving an average value of $3.5\,\mathrm{MN\,m^{-3/2}}$. K_{C2} for the same temperature range was an determined from the two sets of data using Equation 9 and $\sigma_y(T)$ which was separately obtained from tensile experiments. Because of the complications of brittle fracture before yield below $-100°$ C, σ_y was extrapolated down to $-160°$ C and subsequently used in the K_{C2} calculations. This effect, however, has not produced marked changes in the general K_{C2} versus T pattern shown in Fig. 8. It seems that the changes observed at $-80°$ C and $-140°$ C can be associated with corresponding β and γ peaks in the $\tan\delta$ versus T plot obtained by Starkweather [8] and for the same material with 0.2% water content.

Table II shows yield stress, modulus e_y ($= \sigma_y/E$) and computed u values (using Equation 10) obtained from these data. The yield strain increases from 0.033 at 20° C to 0.046 at $-40°$ C and remains roughly constant at 0.05 below $-60°$ C. Unlike PMMA and polycarbonate which have constant crack opening displacements at fracture (1.7 and 37 μm, respectively), u for nylon varies directly with $(K_{C2}/\sigma_y)^2$. For the temperature range shown, the magnitudes of u can vary between 110 μm at $-120°$ C and 290 μm at 0° C. Fig. 9 shows a plot of K_{C2} against σ_y, where the two straight lines passing through the data corresponding to the β and γ processes give the plane strain fracture toughness (K_{C1}) when $\sigma_y \to 0$. Furthermore, if crack opening displacement is analysed according to Equation 11, this gives con-

TABLE II Variation of u and e_y with T for nylon

Temperature T (°C)	Yield stress σ_y ($\mathrm{MN\,m^{-2}}$)	Yield strain e_y	Modulus E ($\mathrm{GN\,m^{-2}}$)	Crack tip opening displacement u (μm)
20	80.0	0.033	2.46	395
0	89.0	0.035	2.51	290
−20	100.0	0.039	2.53	238
−40	116.5	0.046	2.56	200
−60	140.0	0.053	2.62	192
−80	159.0	0.055	3.86	161
−100	180.0*	0.056	3.25	122
−120	197.0*	0.057	3.45	110
−140	212.0*	0.059	3.62	131
−160	220.0*	0.058	3.80	88

*Extrapolated values

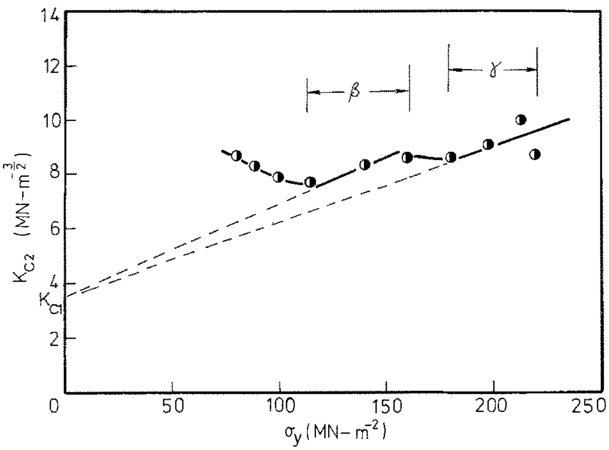

Figure 9 Variation of K_{C2} with yield stress for nylon.

stant u values of 52 and 36 μm for fractures in the β and γ processes, respectively.

6. Conclusion

By using SN and SEN tests, brittle fractures have been produced in both polypropylene and nylon in the temperature range −180 to −10° C and −160 to 20° C, respectively. The differences in K_C obtained from these two sets of data evidently indicated a thickness dependent effect. This phenomenon has been sufficiently explained by the concept of a plane stress K_{C2} and a plane strain K_{C1} toughness value. For both polypropylene and nylon, K_{C1} is almost invariant in temperature, since the material is under essentially plane strain conditions which tend to limit viscoelastic effects. However, K_{C2} determined under plane stress conditions is substantially dependent on shear processes giving large vicsoelastic effects. In polypropylene, K_{C2} increases linearly with σ_y for −10 to −60° C and again for −100 to −180° C. These increases in K_{C2} are associated with viscoelastic energy losses due to the β and γ processes. Using a modified crack tip opening displacement equation (i.e. Equation 11), a constant u of 91 μm was obtained for cracking in the temperature ranges corresponding to these two molecular relaxation processes. K_{C2} values of nylon show changes in those temperature regions corresponding to the tan δ peaks associated with the β and γ processes. This one-to-one correspondence between changes in K_{C2} and peaks in tan δ versus temperature plots has also been reported by Vincent [9] in the impact data for PTFE. Constant crack tip opening displacements were also obtained in nylon for fractures in the β and γ ranges, with values of 52 and 36 μm, respectively.

References

1. G. P. MARSHALL, L. H. COUTTS and J. G. WILLIAMS, *J. Mater. Sci.* 9 (1974) 1409.
2. M. PARVIN and J. G. WILLIAMS, *ibid* **10** (1975) 1883.
3. *Idem, ibid,* **11** (1976) 2045.
4. A. G. ATKINS, C. S. LEE and R. M. CADDELL, *ibid* **10** (1975) 1381.
5. Y. W. MAI and A. G. ATKINS, *ibid* **11** (1976) 677.
6. W. F. BROWN, Jr and J. SRAWLEY, *ASTM STP 410* (1966).
7. G. R. IRWIN, *J. Appl. Mechs.* December 29 (1962) 651.
8. H. W. STARKWEATHER in "Nylon Plastics", edited by M. I. Kohan (John Wiley and Sons, New York, 1973) p. 307.
9. P. I. VINCENT, *Polymer* **15** (1974) 111.

Received 25 October and accepted 22 November 1976.