JOURNAL OF MATERIALS SCIENCE 31 (1996) 291–295

The crack growth resistance of polyimide film

B. COTTERELL
School of Mechanical and Production Engineering, Nanyang Technological University, Singapore 2263
M.C. SIM
Matcor Technology and Services Pte Ltd, 16 Science Park Drive, Singapore 0511
G. AMRUTHARAJ, S.H. TEOH
Department of Mechanical and Production Engineering, National University of Singapore, Singapore 0511

The crack growth resistance of Dupont Kapton film is measured using the constrained short centre-notched tension specimen that is clamped along its loaded edge. Poisson's effect induces a lateral tension on the specimen which eliminates the buckling problems associated with standard fracture tests. Polyimide film is shown to possess considerable crack growth resistance that decreases slightly with film thickness. SEM examination reveals the likely cause of the crack growth resistance to be cold drawing behind the crack tip.

1. Introduction

Polyimide films have the ability to maintain their physical, electrical and mechanical properties over a wide temperature range. These properties make polyimide a useful and unique industrial material and has opened new design and application areas such as electronic and microelectronic components (circuit boards, insulators, flexible cables), automotive components (internal working parts, switches) and medical devices (prostheses, circuitry in bio-implants). However, one limitation to its use is the low toughness of polyimide.

The fracture toughness of Kapton, a Dupont polyimide has been reported by Hinkley and Mings [1]. They used a single-edge notched (SEN) tension specimen to evaluate the initiation fracture toughness. Because the load at fracture was considerably larger than at initiation, and the loading rate was comparatively fast (1.85 mm min^{-1} over a 50 mm specimen), the polyimide film obviously exhibited considerable crack growth resistance. Polyimide is a viscoelastic material and its properties are time-dependent. Popelar *et al.* [2] have determined the crack growth rate of Kapton film as a function of the stress intensity factor, also utilizing the SEN tension specimen. Although Popelar *et al.* [2] did not report a crack growth resistance they noted that for the 100 HN film "a transitory period marked by a decrease of the crack growth rate with increasing crack length and stress intensity factor is apparent", which indicates crack growth resistance. There is considerable discrepancy between the properties reported by Hinkley and Mings [1] and Popelar *et al.* [2]. For 25 μm Kapton film, which is assumed to be 100 HN, Hinkley and Mings [1] report an initiation fracture toughness of 1.65 ± 0.14 MPa m$^{1/2}$, whereas Popelar *et al.* report that for 500 HN Kapton (nominal thickness 125 μm) crack initiation took a long time if the stress intensity factor was less than 6.9 MPa m$^{1/2}$, and for 100 HN film they only gave crack growth data for stress intensity factors greater than 5.3 MPa m$^{1/2}$. One possibility for the large discrepancy between these values is that the SEN geometry used by both causes a free film to buckle and to avoid buckling both used anti-buckling plates. To enable the stress intensity factor at the tip of the crack in a SEN specimen to be determined accurately, the anti-buckling plates have to be very finely adjusted. If the clearance between the plates is too large, buckling occurs, and if too small, there is friction between the plates and the film. Hinkley and Mings [1] did not discuss the clearance used in their tests. Popelar *et al.* [2] used Mylar tape on the anti-buckling plates to reduce friction and stated that for the 100 HN film the clearance was 50 μm. The problem is that the standard fracture test geometries are not really suited for testing thin films because of the compressive stresses parallel to the notch which cause buckling in thin films.

Recently Tielking [3] has used a constrained short tension (CST) specimen that makes use of Poisson's effect to induce a transverse tensile stress that helps to prevent buckling so that thin materials can be tested without the necessity of using restraining plates. The specimen is a centrally notched strip whose edges are clamped between grips which prevent lateral contraction and thus induce a transverse tensile stress, when the strip is stretched, preventing buckling (see Fig. 1). Tielking [3] used this geometry to obtain J_R-curves for polyethylene films. The CST specimen is also eminently suitable for testing the more brittle polymeric films whose fracture characteristics can be described by linear elastic fracture mechanics [4]. The

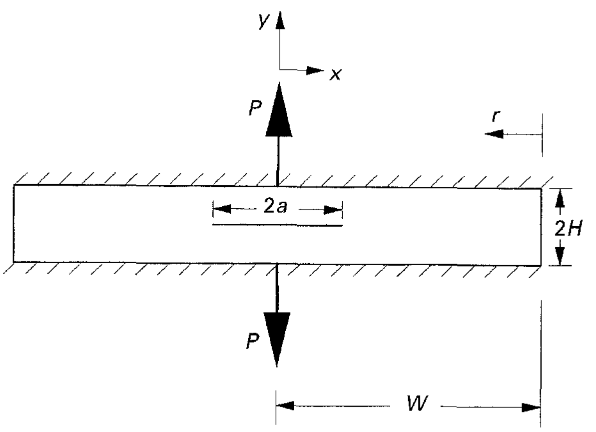

Figure 1 Schematic representation of the constrained short tension specimen.

short-time crack growth resistance of Kapton film obtained with the CST test specimen is described in this paper.

2. Experimental procedure

The essential features of the equipment (Fig. 2) for a CST test are grips of sufficient bending stiffness so that the deflection of the grips under load is minimal. The grips must have faces that prevent the material from slipping. For these tests on polyimide film, 120 grade silicon carbide paper was used to face the grips, but silicon rubber facings, as suggested by Tielking [3], would be equally suitable. The grips are housed in a heavy C-shaped fixture which has high bending stiffness and tightened on to the film by six set screws. The width of the biaxial specimen was 210 mm and the height, $2H$, was 30 mm.

3. Crack growth resistance tests

Dupont Kapton polyimide film 100 HN, 200 HN, 300 HN, and 500 HN were the test materials. The measured thickness of these films, which differed slightly from the nominal thicknesses, are given in Table I. Central notches of total length of 19.5–26.5 mm were cut, perpendicular to the machine direction, with a surgical knife to obtain a sharp tip. The crack tip (see Fig. 3) is as sharp as that obtained with a razor using a slicing action [1,2]. Half-crack lengths varying from 9.5–15.5 mm were used in the tests. A paper backing was used to support the films while they were mounted in the grips. This paper was cut prior to loading the specimens at a strain rate of $1\,\mathrm{mm\,min^{-1}}$ at a temperature of 22 °C in a J.J. Loyld M30K tensile testing machine. Photographs of the crack were taken at timed intervals from which the crack extension could be related to the load. The Poisson's ratio is assumed to be 0.35 as stated by the manufacturer.

For cracks whose length is longer than 0.8 of the specimen height, the stress intensity factor is given by the approximate relationship [4]

$$\frac{K}{\sigma[H(1-\nu^2)]^{1/2}} = 1\bigg/\left(C - \frac{a}{W}\right) \qquad (1)$$

where σ is the gross stress, ν Poisson's ratio, H the half-height, W the half-width, and $C = 1 + (0.3154 - 0.7666\,\nu^2)(H/W)$. However, here the stress intensity

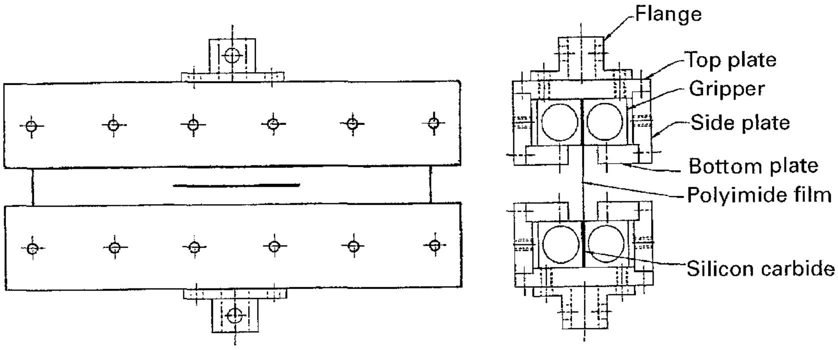

Figure 2 Details of the constrained short tension specimen used to test polyimide film.

TABLE I Crack growth resistance of Kapton

	Film type			
	100 HN	200 HN	300 HN	500 HN
Film thickness (μm)	28	53	79	129
Initiation toughness ($\mathrm{MPa\,m^{1/2}}$)	2.88	3.54	2.96	2.40
Plateau toughness ($\mathrm{MPa\,m^{1/2}}$)	5.23	5.04	4.66	3.74
Crack growth to 90% K_∞ (mm)	1.3	2.0	2.6	3.0
Crack growth to 95% K_∞ (mm)	1.9	3.2	4.0	4.6
Max. width of isochromatics (mm)	0.12	0.23	0.34	0.36
Crack growth to max (mm) width of isochromatics	0.6	1.4	1.8	1.9

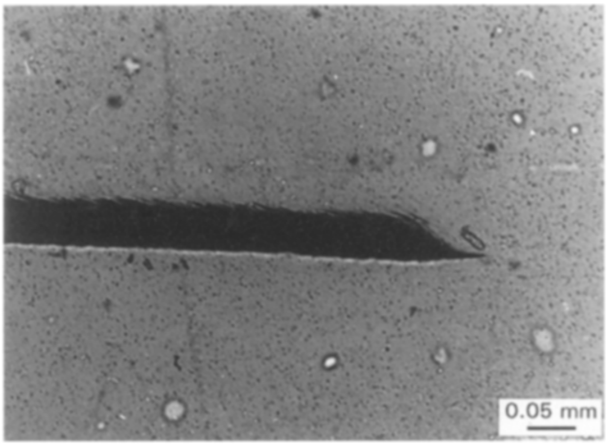

Figure 3 Tip of a crack cut with a surgical knife.

factor has been calculated directly from a finite element solution as previously described [4].

4. The crack growth resistance of Kapton polyimide film

The stress intensity factors are plotted against the crack extension to give the K_R-curves shown in Figs 4–7. The results are independent of the initial crack length. The radius, r_Y, of the plastic zone was estimated, using Irwin's equation [5]

$$r_Y = \frac{1}{2\pi}\left(\frac{K_{\max}}{\sigma_Y}\right)^2 \tag{2}$$

to be less than 1 mm. Thus the suggested criterion for validity [4]

$$H > 14r_Y \tag{3}$$

is satisfied and the application of linear elastic fracture mechanics is justified. Correction for the size of the plastic zone is negligible. The exponential curve gives a good fit to the crack growth resistance and these curves are compared in Fig. 8 for the four film thicknesses. The exponential curves have also been used to obtain the plateau values of the crack growth resistance shown in Fig. 9. The initiation fracture toughnesses have been obtained by fitting the curve

$$K_R = A + B\Delta a^n \tag{4}$$

for $\Delta a < 1.5$ mm, which give a better fit than the slightly different method recommended in ASTM E 1290, and are shown in Fig. 9.

5. Discussion of the crack growth resistance

The K_R-curves show that the polyimide films possess considerable crack growth resistance with a range of the order of 2 mm and the plateau fracture toughness is sensitive to the film thickness with the thinnest film displaying the highest toughness. The initiation values of the fracture toughness are more difficult to estimate accurately. For Kapton 100 HN, the initiation fracture toughness, 2.88 MPa m$^{1/2}$, is significantly larger than the toughness (1.65 ± 0.14MPa m$^{1/2}$) reported by Hinkley and Mings [1]. However, the initiation fracture

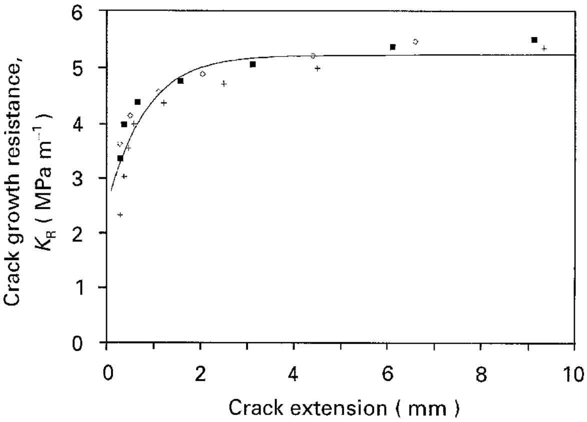

Figure 4 Crack growth resistance curve for Kapton 100 HN polyimide film; symbols, experimental data; (——) exponential curve of best fit.

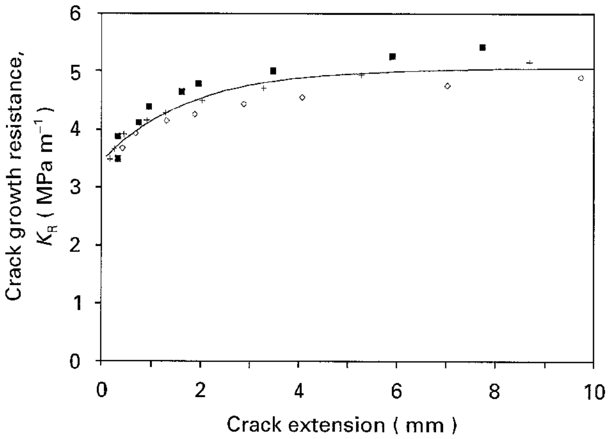

Figure 5 Crack growth resistance curve for Kapton 200 HN polyimide film; symbols, experimental data; (——) exponential curve of best fit.

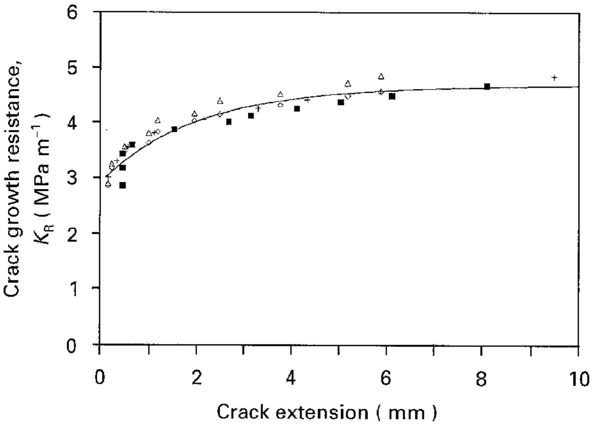

Figure 6 Crack growth resistance curve for Kapton 300 HN polyimide film; symbols, experimental data; (——) exponential curve of best fit.

toughness of 2.96 MPa m$^{1/2}$ for Kapton 300 HN is smaller than the toughness (≈ 6.9 MPa m$^{1/2}$) inferred from the work of Popelar *et al.* [2]. It is suspected that the reasons for these discrepancies, which are not systematic, may lie in the test method. The plateau values of the crack growth resistance decreases with film thickness as does the time-dependent crack growth rate [2]. However, the

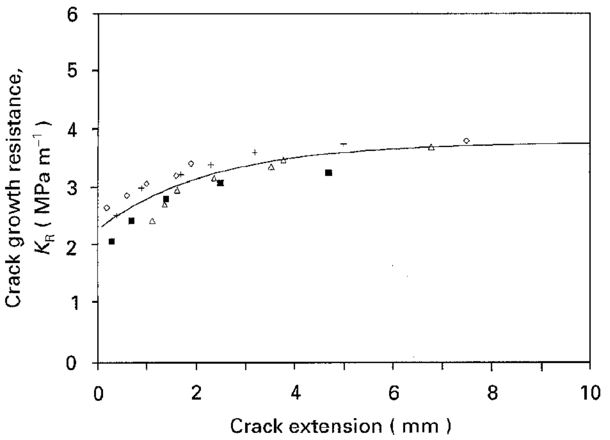

Figure 7 Crack growth resistance curve for Kapton 500 HN polyimide film; symbols, experimental data; (——) exponential curve of best fit.

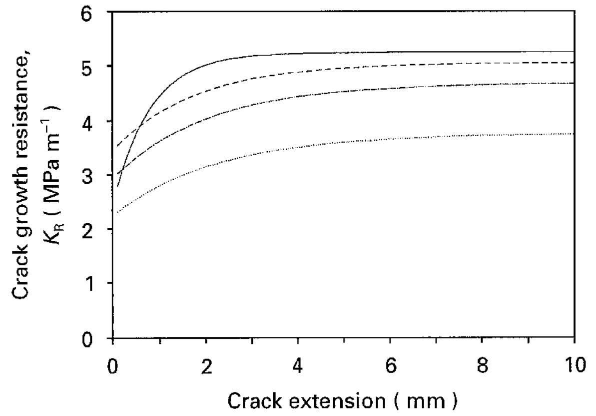

Figure 8 Comparison of the crack growth resistance curves for Kapton polyimide films: (——) 100 HN, (– – –) 200 HN, (–·–) 300 HN, (···) 500 HN.

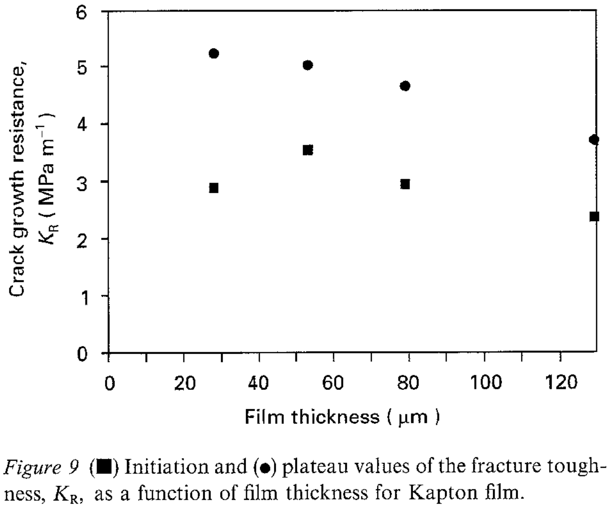

Figure 9 (■) Initiation and (●) plateau values of the fracture toughness, K_R, as a function of film thickness for Kapton film.

picture is less clear for the initiation fracture toughness. It appears (see Fig. 9) that the initiation toughness is a maximum for Kapton 200 HN. The crack extension necessary to reach the plateau of the crack growth resistance increases with the film thickness (see Fig. 10) and fits a curve that indicates that the film loses its crack growth resistance as the thickness tends to zero.

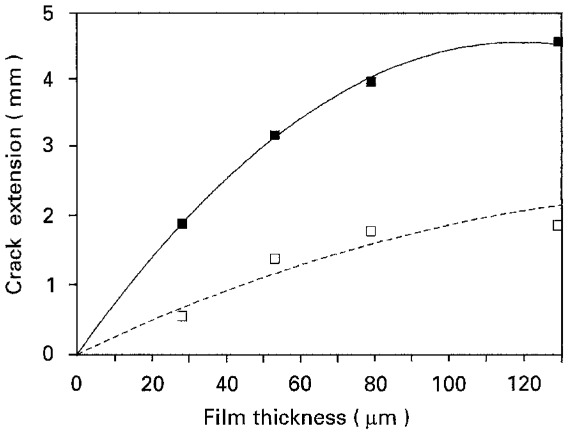

Figure 10 The crack growth resistance range (—■—) Extension to 95% K, (– –□– –) extension to maximum isochromatic width.

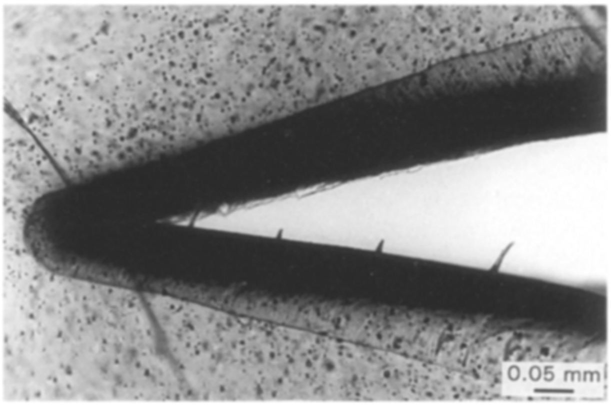

Figure 11 Cold drawing in a Kapton 200 HN film.

Photographs (see Fig. 11) reveal evidence of cold drawing behind the crack tip and also show a small darker zone adjacent to the fracture edge where the film seems to be necked. Scanning electron micrographs also show necking of the film at the initiating notch and reveal delamination on one or more planes (Fig. 12a). Away from the site of fracture initiation, the remnants of cold drawing can be clearly seen (Fig. 12b). Because the initiation and plateau values of the crack growth resistance do not tend to the same value as the film thickness tends to zero (see Fig. 9), it must be assumed that cold drawing still would take place in very thin films, but that it would be confined to a very small region behind the crack tip.

The fractured surfaces were examined under polarized light to study the plastically deformed zone left behind the crack tip process zone. The colours of the isochromatics observed showed increasing orders towards the fracture edge. The width of the isochromatic band increases with the film thickness (see Table I). However, that does not necessarily mean that the width of the plastic zone also increases with film thickness, because the isochromatic fringe order is proportional to the film thickness. The distance to achieve the maximum width of the isochromatics is less than the distance to achieve the plateau value of the fracture toughness but increases with film

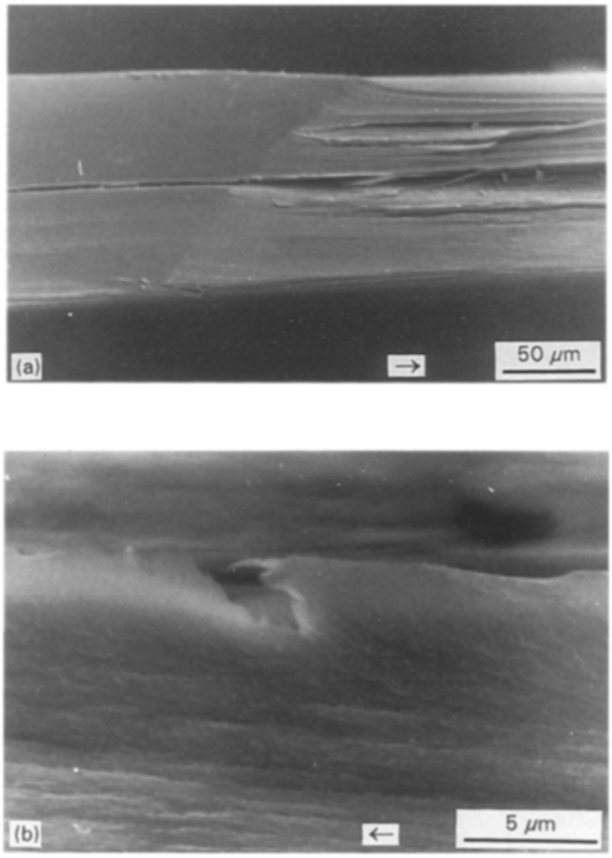

Figure 12 Scanning electron micrographs of the fracture surface of Kapton 500 HN film (the arrows show the propagation direction), (a) at the initiation point, (b) away from the initiation point.

thickness, as does the crack extension necessary to achieve the plateau toughness (see Fig. 10).

A probable mechanism of the crack growth resistance is cold drawing of the polyimide behind the crack tip. This drawing action will absorb increasing energy as the crack extends and the drawing is left in the wake of the crack tip and hence the fracture toughness increases with crack extension. The cold drawing will be time-dependent and will contribute to the time-dependent crack growth observed by Popelar *et al.* [2].

6. Conclusions

The fracture toughness of polyimide film can be conveniently measured using the constrained short tension specimen. This geometry eliminates buckling which is a problem with the standard test geometries. Because the plastic zone is small, linear elastic fracture mechanics can be used to describe the behaviour of polyimide film. The initiation fracture toughness measured in this study is larger than that reported by Hinkley and Mings [1], but smaller than that inferred from the work of Popelar *et al.* [2]. It is suspected that inaccuracies could arise from the use of the SEN specimen by these researchers because of buckling problems.

There is considerable crack growth resistance, over a range of about 2 mm, which decreases slightly with film thickness. SEM shows that the probable mechanism for the crack growth resistance is micro-drawing of the polyimide behind the crack front.

Acknowledgements

The supply of Kapton film by Dupont Singapore Pte Ltd is gratefully acknowledged. Mr Philip Phua of Dupont Singapore is also warmly thanked for his encouragement and help.

References

1. J. A. HINKLEY and S. L. MINGS, *Polymer* **31** (1990) 75.
2. S. F. POPELAR, C. H. POPELAR and V. H. KENNAR *J. Electron. Packaging* **115** (1993) 264.
3. J. T. TIELKING, *Polym. Test.* **12** (1993) 207.
4. B. COTTERELL, M. C. SIM, G. AMRUTHARAJ and S. H. TEOH, *J. Test. Eval.* to be published.
5. G. R. IRWIN, In: Structural Mechanics, Proceedings of the first symposium on Naval Structures (1960) 557.

Received 7 June
and accepted 22 June 1995