JOURNAL OF MATERIALS SCIENCE 26 (1991) 1387–1390

Temperature effects on quasi-static fracture of PMMA

M. BALZANO, K. RAVI-CHANDAR
Department of Mechanical Engineering, University of Houston, Texas, USA

The effect of temperature on the quasi-static fracture behaviour of PMMA is examined. It is found that the critical stress intensity factor, K_{IO}, at crack growth initiation decreases with temperature up to a certain critical temperature, T_{c}. Above T_{c}, K_{IO} increases rapidly. The speeds with which slow crack growth could be maintained without transition to brittle fracture were found to be significantly higher at high temperatures.

1. Introduction

The mechanical behaviour of polymeric materials near ambient temperatures is very sensitive to temperature because the softening temperature of most of these materials is very close to ambient temperatures. In particular, the fracture behaviour of these materials could undergo vast changes from low-energy brittle failure to ductile tearing with large energy dissipation. Such transitions also occur in metallic materials at the nil-ductility-temperature as observed in Charpy tests. Brittle-to-ductile transitions have also been reported in polymers like polycarbonate. This type of a transition with the attendant increase in fracture energy has important applications in polymer matrix composite materials, particularly those with a thermoplastic matrix. In this paper we look at the fracture behaviour of polymethylmethacrylate (PMMA) as a function of temperature and loading rate.

A vast literature exists on the fracture behaviour of PMMA where much attention has been focused on the craze-dominated fracture behaviour and the characterization of the stress intensity factor–crack speed relationship. For example, Döll [1] has studied the crazing phenomenon and its influence on fracture extensively. Marshall *et al.* [2] looked into the temperature effects on the stress intensity factor–crack speed relationship. They also investigated the effect of temperature on the crack initiation and instability behaviour and report that the stress intensity factor at initiation is a maximum at around − 80 °C (2.4 MPa $\mathrm{m}^{1/2}$) and drops slowly to about 1.7 MPa $\mathrm{m}^{1/2}$) at colder temperatures. The critical stress intensity factor also drops at higher temperatures, dropping to about 0.8 MPa $\mathrm{m}^{1/2}$ at around 80 °C. Some difference was noted between the stress intensity factors at initiation of slow crack growth and at transition to fast crack growth as temperature increased, but below − 60 °C, no slow crack growth was observed and only brittle crack growth was reported. The present paper looks into the various regimes of crack initiation behaviour as a function of the temperature. Also the effects of temperature on slow crack growth and transition to fast crack growth are examined. In Section 2, we briefly describe the experimental procedure used in these experiments. The effect of temperature on crack initiation is discussed in Section 3 and finally, the effect of temperature on slow crack growth and transition to fast crack growth are examined in Section 4. These latter results are used to support a mechanism proposed earlier [3] for ductile to brittle transition observed in PMMA.

2. Experimental procedure

The details of the experimental procedure are described elsewhere [3] and only a brief sketch is provided here for the sake of completeness. Single-edge notched (SEN) specimens were used in the experiments. 200 μm wide slits were laser machined into the specimen and a sharp crack tip was obtained by hammer impact of a razor blade inserted into the slit. The specimens were pulled to fracture in an Instron tensile testing machine at constant crosshead rates ranging from 0.25–500 mm min^{-1}. A furnace was specially built for the temperature control in the range − 60–150 °C, with double glass windows for the implementation of the optical method of caustics, because, in addition to the load measurements, the crack tip stress intensity factor was determined directly by the method of caustics. This technique has become quite popular in recent years and a summary of its application can be found in [4]. Briefly, the formation of the caustic may be explained as follows: a light ray passing through the near tip region of a cracked specimen is deviated away from its straight path due to the thickness gradient and also due to the change in the refractive index caused by the stress-optic effect. If the light beam is viewed on a screen plane at some distance behind the specimen, a dark region called the shadowspot, surrounded by a bright curve called the caustic curve is observed. From the size of the shadowspot, the stress intensity factor can be obtained [4]. The crack tip itself is obscured by the shadowspot and its location has to be determined from the geometry of the shadowspot. In the present experiments,

0022–2461/91 $03.00 + .12 © *1991 Chapman and Hall Ltd.*

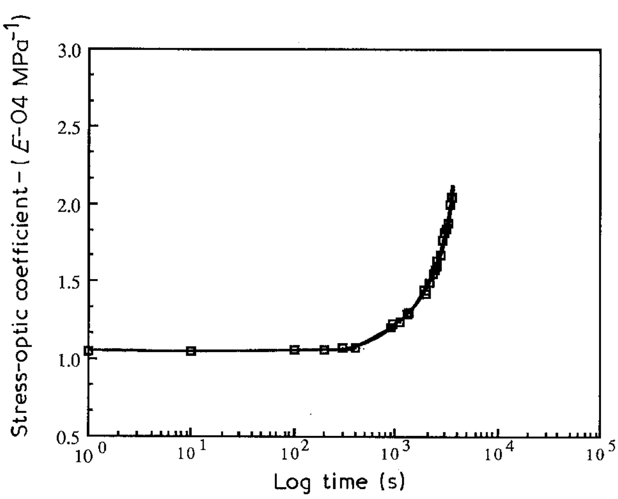

Figure 1 Time dependence of the stress-optic coefficient for PMMA.

because the temperature was varied over the range -60–150 °C, the method of caustics has to be extended to include viscoelastic effects. Kim *et al.* [5] addressed this problem by constructing a master curve for the stress-optic coefficient from relaxation experiments. We adopted this approach and obtained the master curve for the stress-optic coefficient for PMMA, as shown in Fig. 1. However, because each experiment was conducted at a fixed temperature, over very short times, the relationship between the caustic diameter and the stress intensity factor could have been obtained by simple calibration.

The optical apparatus was set up around the testing machine and the shadowspot formed on the screen plane was captured using a high-speed video system capable of scanning at 12000 frames s^{-1}. From this video record, the variation of the stress intensity factor with time as well as the crack position was determined. From the time histories of the stress intensity factors and crack position, the instant of crack initiation as well as the critical stress intensity factor at crack growth initiation, $K_{\mathrm{I}0}$, were found through interpolation.

3. Temperature effects on crack initiation

At room temperature the value of $K_{\mathrm{I}0}$ was found to be nearly constant at 1.08 MPa m$^{1/2}$ for PMMA (independent of the rate of loading in the range of $K'_{\mathrm{I}0} =$ 0.02–10 MPa m$^{1/2}$ s^{-1}). These results are in contrast to the results obtained by Marshall *et al.* [2] who found (from load measurements) that the stress intensity factor at initiation increased from about 0.7–1.0 MPa m$^{1/2}$ as the loading rates increased in the same range used in the present work. It is possible that the difference is due to the difference in the materials because the properties of PMMA are dependent on the molecular weight [1]. It is also possible that the differences in the way the initial cracks were introduced (which cause residual stresses to be set up near the crack tip) caused this difference in the result.

Experiments were performed at temperatures ranging from -55–120 °C, at a fixed rate of loading on PMMA. An overall view of the variation of $K_{\mathrm{I}0}$ with temperature for a crosshead rate of 5 mm min^{-1} is shown in Fig. 2. The critical stress intensity factor decreases with increasing temperatures, from 1.75 MPa m$^{1/2}$ at -55 °C to 0.6 MPa m$^{1/2}$ at 80 °C where a minimum is reached as illustrated in Fig. 2. We refer to this temperature at which a minimum critical stress intensity factor is observed as T_c. Also a noticeable plateau in the region -10–0 °C is observed. Above 80 °C, $K_{\mathrm{I}0}$ increases with increasing temperature to reach 1.06 MPa m$^{1/2}$ at 95 °C. Finally, above this temperature, large deformation of the specimen occurs and initiation of crack is not observed; distributed crazing occurs at the crack tip area. Although these results present some small differences in the values of the stress intensity factors at initiation compared to those of Marshall *et al.* [2], the general trend is the same; i.e. increasing $K_{\mathrm{I}0}$ with decreasing temperatures. However, the minimum value of $K_{\mathrm{I}0}$ at T_c was not observed in [2]. The experimental observations are discussed below, based on the ability of the material to be drawn into craze.

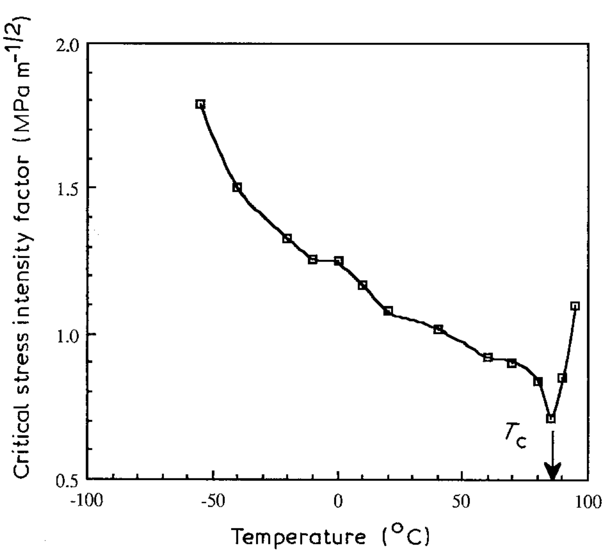

Figure 2 Temperature dependence of the critical stress intensity factor for PMMA.

It is well known that the mechanism of crack initiation for PMMA, in the range of temperatures and loading rates investigated, is governed by the formation of a single dominant craze at the crack tip [1]. As the temperature is decreased, it becomes more difficult to draw crazes. At the limit, no craze is initiated (for instance PMMA in the SEN configuration at -20 °C for a loading rate of 5 mm min^{-1}) and only brittle fracture is initiated. If the temperature were to be decreased further, a plateau of maximum value for $K_{\mathrm{I}0}$ should be reached, because once craze formation is inhibited, the effects of decreasing the temperature should diminish. Moreover for very low temperatures, $K_{\mathrm{I}0}$ should even decrease due to the fact that the material becomes more and more brittle giving a lower value of the fracture toughness. We did not observe this plateau or the drop in $K_{\mathrm{I}0}$ (temperatures were not low enough) but Marshall *et al.* [2] reported experiments on PMMA at 5 mm min^{-1} where $K_{\mathrm{I}0}$ reached a maximum around 2.0 MPa m$^{1/2}$ between -80 and -100 °C and dropped to 1.6 MPa m$^{1/2}$ at

− 180 °C. Marshall *et al.* [2] also observed the plateau at around 0 °C and attributed to the effect of freezing of absorbed moisture.

On the other hand, increasing the temperature closer to the softening temperature of the material allows easier drawing of the material into craze, leading to lower critical stress intensity factor for initiation of crack growth. But this is true only up to a certain temperature, T_c. Beyond this critical temperature, the critical stress intensity factor increases with increasing temperature. It appears that above T_c the crack tip starts to blunt and it is known that the stress concentration is much higher in a sharp crack tip than in a blunted one. Thus the specimen with the blunted crack tip will withstand higher loads, leading to an increase in the apparent material toughness. Under such conditions, fast crack growth is initiated instead of the slow crack growth observed at temperatures below T_c. Finally, for very high temperatures the crack tip is not a weak point any more. Large, irreversible deformation of the sample occurs with distributed crazing all over the specimen. An example of this distributed crazing is shown in Fig. 3. It is interesting to note that the crazes are formed along specific orientations in front of the crack tip. These directions appear to be aligned along the directions of the maximum principal directions as obtained from the singular stress field around the crack tip. Also, in the immediate vicinity ahead of the crack tip itself, extensive crazing is not observed due to the fact that the dominant crack tip craze, as well as the surrounding craze field, effectively unload this region (shielding effect). Also, the high biaxiality of the stress field in front of the crack tip inhibits craze formation in this region. The distributed crazes form only as a surface layer and do not penetrate into the thickness of the plate.

In conclusion, as a function of the temperature, different mechanisms of crack growth have been observed. At low temperatures, brittle crack growth, through microcrack nucleation and coalescence, is initiated. The second crack initiation mechanism is due to craze formation at the crack tip and has been mainly observed in the temperature range 20 °C to T_c. This leads to slow crack growth with velocities in the range 0.01–1 $\mathrm{m\,s^{-1}}$. Thirdly, due to large deformations at temperatures greater than T_c, blunting dominates the fracture process. Finally, at temperatures higher than 95 °C, fracture does not occur due to crack tip processes; large deformation of the specimen with distributed crazing predominates. Thus low rate deformations at around room temperature or higher lead to the most critical conditions for crack initiation in PMMA. Similar behaviour is expected in other thermoplastic polymers that are capable of crazing. Temperature dependence similar to that shown in Fig. 2 was reported by Wang [6] for PEEK (polyetheretherketone).

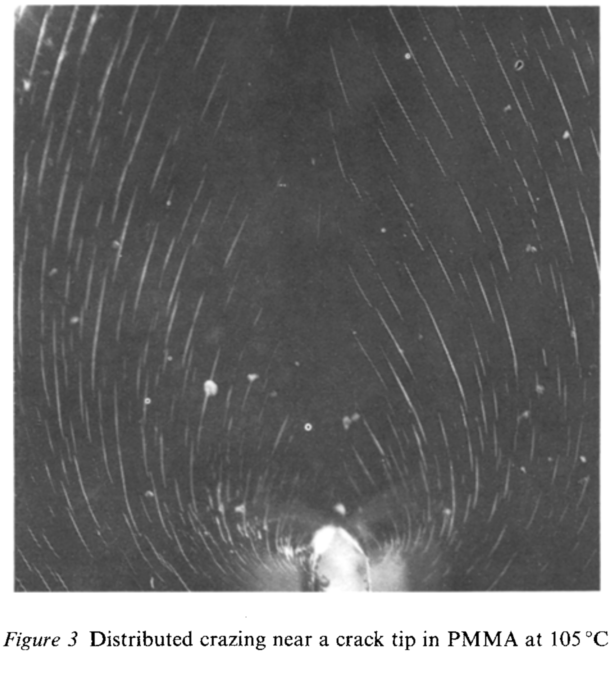

Figure 3 Distributed crazing near a crack tip in PMMA at 105 °C.

4. Temperature effects on slow crack growth and transition to fast crack growth

Once a crack has been initiated into growth, the nature and extent of further crack growth depends on the temperature, rate of loading, as well as the type of specimen. For example, at room temperature, at a rate of loading of about 0.02 $\mathrm{MPa\,m^{1/2}\,s^{-1}}$, slow crack growth is initiated; the speed of the crack is of the order of several $\mathrm{mm\,s^{-1}}$. Upon continued loading through crosshead motion at a constant rate, an abrupt change occurs in the crack speed; the speed changes to several hundred $\mathrm{m\,s^{-1}}$. This behaviour has been labelled as transition and has been discussed extensively in the literature. Transition has been attributed to several causes such as β_r-relaxation peak [7], isothermal-to-adiabatic transition [2] and ductile-to-brittle transition [3]. Through an investigation of the molecular weight dependence of the transition speed, Döll [8] discounted the possibility of the β_r-relaxation peak model. The isothermal–adiabatic transition model implies that the velocity at transition should only be weakly dependent on temperature (because the diffusion properties of PMMA are only weakly dependent on the temperature in the range 0–100 °C); this will be examined later. A ductile-to-brittle transition model was proposed earlier [3] to account for the observed crack speed jump; the model is briefly summarized here. It was postulated that slow crack growth was essentially governed by a dominant crack-tip craze and fast crack growth was through microcrack nucleation and coalescence. As a result of the diffusion of the energy released from the crack tip processes, the temperature in the crack tip region increases; this facilitates faster crack growth because the material fracture toughness decreases as temperature increases. However, as the crack speed increases further, diffusion is limited and the temperature ahead of the crack tip does not increase appreciably; brittle fracture occurs with the crack propagating at very high speeds. The temperature field can, in fact, be evaluated from the calculations of Weichert and Schönert [9]; their results indicate that at higher crack speeds the region ahead of the crack tip, in which the temperature increases above the ambient, becomes

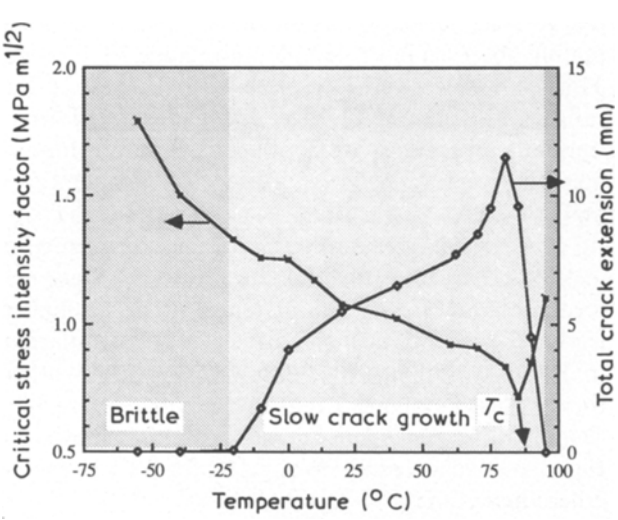

Figure 4 Temperature dependence of slow crack growth.

smaller. Based on this model it can be predicted that by increasing the ambient temperature, it would be possible to eliminate the ductile-to-brittle transition altogether. We now present some results on this aspect of crack growth behaviour. Fig. 4 shows the amount of slow crack growth as a function of temperature, as well as the critical stress intensity factor for crack initiation; the velocity at transition is plotted against the specimen temperature in Fig. 5.

Slow crack growth is governed by a craze formation at the crack tip. By decreasing the temperature one should be able to inhibit craze formation and observe only brittle fast crack growth. As indicated in Fig. 4, at temperatures below $-20\,^{\circ}$C, slow crack growth is not observed and crack initiation is of a brittle nature. On the other hand, increasing the temperature facilitates slow crack growth, thus delaying transition to fast crack growth. This has been verified experimentally; as indicated in Fig. 4, the total extent of slow crack growth before transition to fast crack growth increases with increase in temperature, indicating a delay in transition. The critical crack speed at transition also increases; this is a reflection of the fact that it is easier to propagate a crack in the thermally softened material (K_{10} is smaller at higher ambient temperature). This increase in crack speed with temperature clearly indicates that transition has been delayed. This lends support to the contention [3] that increasing the ambient temperature facilitates craze formation thereby leading to slow craze-dominated crack growth without transition. The isothermal–adiabatic transition model of [2] indicates a lower bound for critical speed at transition, which is also displayed in Fig. 5. While there is some correspondence between the isothermal–adiabatic transition model and the experimentally determined critical speeds for temperatures below 60 °C, there is a large discrepancy at higher temperatures.

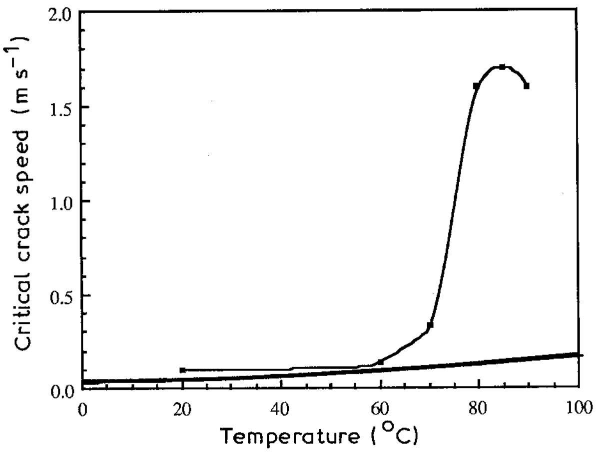

Figure 5 Temperature dependence of (-■-) the critical speed at transition to fast crack growth. (——) from [2].

5. Conclusions

The temperature dependence of fracture in PMMA was investigated. It was found that the critical stress intensity factor necessary to initiate crack growth decreased with increasing temperature; a minimum value of the fracture toughness was observed at a critical temperature labelled T_c. The crack growth was brittle at temperatures below $-20\,^{\circ}$C; slow crack growth with increasing crack speeds at transition was observed below 90 °C and crack growth could not be initiated above 95 °C.

Acknowledgement

The research support from the National Science Foundation, Solid Mechanics Program (Grant no. MSM-8415244) is gratefully acknowledged.

References

1. W. DÖLL, in "Advances in Polymer Science", edited by H. H. Kausch Vol. 52/53 (Springer, Berlin, 1983) pp 105–168.
2. G. P. MARSHALL, L. H. COUTTS and J. G. WILLIAMS, *J. Mater. Sci.* **9** (1974) 1409.
3. K. RAVI-CHANDAR and M. BALZANO, *Engng Fract. Mech.* **30** (1988) 713.
4. J. F. KALTHOFF, in "Handbook of Experimental Mechanics", edited by A. S. Kobayashi (Prentice-Hall, Englewood Cliffs, NJ 1987) pp 430–500.
5. K. S. KIM, K. L. DICKERSON and W. G. KNAUSS, *Int. J. Fract.* **32** (1987) 265.
6. S. S. WANG, "Elevated temperature transverse fracture of thermoplastic matrix composite materials," presented at the ASME Winter Annual Meeting, Chicago (1988).
7. F. A. JOHNSON and J. C. RADON, *Engng Fract. Mech.* **4** (1972) 555.
8. W. DÖLL, *J. Mater. Sci. Lett.* **11** (1976) 2348.
9. R. WEICHERT and K. SCHÖNERT, *J. Mech. Phys. Solids* **22** (1974) 127.

Received 11 April
and accepted 29 November 1989