Direct measurements of the strain on the boundary of crazes in polyethylene

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(Received 20 July 1987; accepted 16 September 1987)

The strain distribution was measured on the boundary of crazes in polyethylene by a new technique where the crazes were produced under plane strain conditions. The strain distribution indicated that crazes in a linear homopolymer are weaker than those in a copolymer. The stress–strain curves of these polymers were used to obtain information about the stress field associated with the observed strain field. These results represent the first direct measurements of the strain field on the boundary of the craze without recourse to theory or arbitrary assumptions.

(Keywords: polyethylene; crazing; stress distribution)

INTRODUCTION

Crazes are the usual precursors for fracture in polymers. In order to understand the initiation and kinetics of fracture, it is very important to know the stress distribution on the boundary between the craze and the matrix. Many investigations have been devoted to determining the stress distribution on the boundary of damage zones that emanate from a crack where the damage may be in the form of a craze, an array of voids, or a plastic zone. The approaches have ranged from the purely theoretical to the partly theoretical and partly experimental. One of the simplest and most useful theories is by Dugdale[1], who assumed that the stress field was constant along the boundary and equal to the yield point. Knight[2], Verheulpen-Heymans and Bauwens[3], Wilczynski *et al.*[4], Walton and Weitsman[5], Bevan[6], Warren[7] and Wilkinson and Vitek[8] used theoretical methods based on artificial assumptions concerning the nature of the damage zone. Brown and Ward[9], Döll[10] and Wang and Kramer[11] measured the geometry of the boundary and from that calculated the stress field. Brown and Ward[9] and Döll[10] used an interferometric technique to measure the shape of the boundary in poly(methyl methacrylate) (PMMA) and Kramer[12] measured the geometry of the boundary in thin films using transmission electron microscopy (TEM). All the approaches have assumed that the matrix is elastic in order conveniently to carry out the necessary calculations. Brown and Ward[9] and Döll[10] found agreement with the Dugdale theory[1]. Imai and Ward[13] found that the Dugdale theory did not explain some of their fatigue results on PMMA. Kramer[12] found that the stress field varied depending on the polymer, but in most cases found a maximum of the stress at the craze tip. In this paper, the strain field on the boundary of crazes in polyethylene (PE) have been measured directly. Information about the stress field may then be derived from the experimentally determined non-linear stress–strain curve for the polymer.

This paper is unique in several aspects: (1) it is the first determination of the strain field on the boundary of a craze where the input is completely based on direct measurements; (2) there are no assumptions about the nature of the craze or the matrix; and (3) the crazes were produced under plane strain conditions in an opaque polymer.

In this paper, the emphasis is on the experimental aspects of the method. In a following paper, this method and the semi-empirical approach of Wang and Kramer[11] will be compared.

EXPERIMENTAL PROCEDURE

Two types of linear PE were investigated: (1) a homopolymer (HPE), with $M_n = 19\,600$ and $M_w = 130\,000$; and (2) an ethylene–hexene copolymer (CPE), with 4.5 butyl chains per 10^3 carbons, $M_n = 15\,000$ and $M_w = 170\,000$. The resins were compression moulded and very slowly cooled to room temperature. Notch tensile specimens were produced whose geometries are shown in *Figure 1*. The notched specimens were exposed to a tensile stress for a given period of time. The damaged zone that emanated from the notch was small compared to the dimensions of the specimen. The conditions of deformation were plane strain. An important difference between the two materials is that growth rate of the damage zone for the HPE is about 10^2 times faster than for the CPE[14]. Thus, the HPE is used for milk bottles and the CPE for gas pipes.

After a certain time, a specimen was unloaded and slices about 1–2 mm thick were taken from the centre of the specimen. The slicing was done with a fresh razor blade at a very slow controlled rate. The slice was then put in a small tensile jig (*Figure 2*) where, under a light microscope, the notch was opened by a definite amount relative to the amount of crack opening displacement (*COD*) that was observed prior to unloading the specimen for slicing. The slice was then coated with gold for the scanning electron microscopy (SEM). An SEM picture is shown in *Figure 3*. Note the parallel scratches that were

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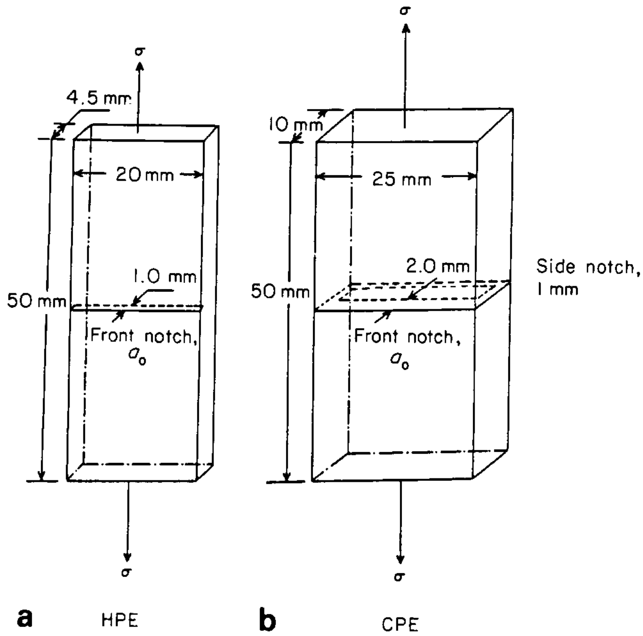

Figure 1 The geometries of the notched specimens of (a) the homopolymer and (b) the copolymer

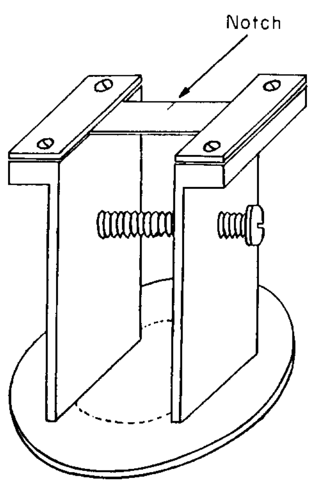

Figure 2 Small tensile jig for SEM

produced by the razor blade when the specimen was sliced. Before loading the slice, the scratches were all parallel. After loading the slice, the spacing of the scratches varied depending on the amount of load.

The SEM photograph was mounted on a wall and the spacing of the razor blade scratches was measured as a function of position by a low-power microscope with a filar eyepiece. Pairs of scratches near the boundary whose spacing was about 0.02 mm apart were chosen for the measurements. The change in spacing of the pair of scratches could be measured with an accuracy of $\pm 0.5\%$. The reference point for zero strain was at a point near the boundary of the free surface of the notch where the stress is zero.

RESULTS

Figure 4 shows the strain distribution along the boundary of the damaged zone in the HPE shown in *Figure 3*. The damage zone started from a notch 1.0 mm deep and was exposed to a stress of 7 MPa at 42°C for 12.5 min. Before unloading for slicing, the opening of the notch at the surface was 126 μm and the crack opening displacement at the notch tip (*COD*) was 24 μm. After slicing, the 2 mm thick slice was loaded so that the *COD* equalled 46 μm. The spacing between pairs of parallel scratches was measured at a series of points 25 μm and 47 μm from the boundary of the damage zone. The strain at each point was calculated by taking the spacing at a point of zero stress position as the gauge length.

The most interesting aspects of the strain distribution are that the maximum stress is at the craze tip and that the stress falls to zero at the notch tip. Far from the craze tip, the strain becomes equal to the value corresponding to the far-field uniform stress.

Figure 5 shows a clearer picture of the structure of the craze that was used for *Figure 4*. The curve in *Figure 4* shows that the boundary stress decreases rapidly in going from the craze tip to the notch tip. It is not possible to make a quantitative judgement about the local strength of the craze by viewing the SEM micrograph. However, the number of broken fibrils at a particular point along the

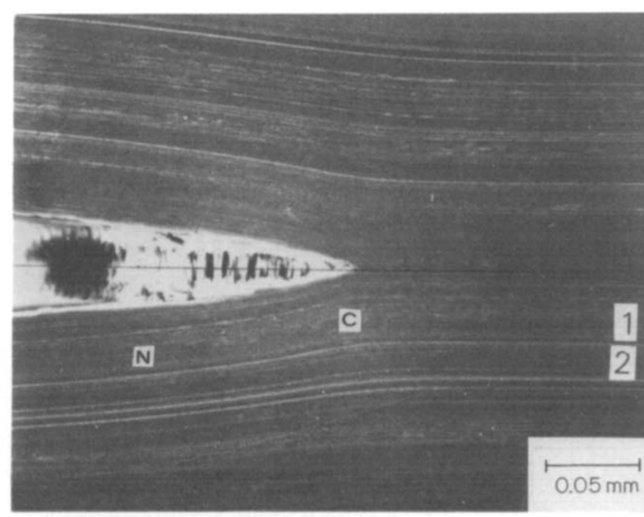

Figure 3 SEM picture of the homopolymer used for measuring strain field (N is notch tip, C is craze tip)

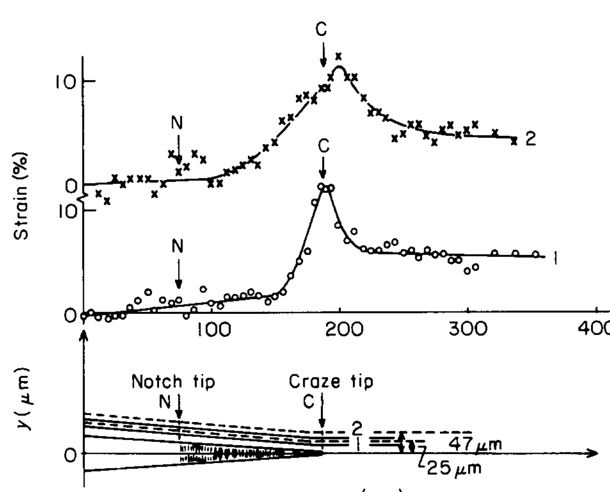

Figure 4 Strain distribution along the damage zone in the homopolymer. Notch depth = 2 mm; $\sigma_1 = 7$ MPa; $T = 42.2$°C; $t = 12.5$ min; $CMOD = 126$ μm, initial $COD = 24$ μm, final $COD = 46$ μm

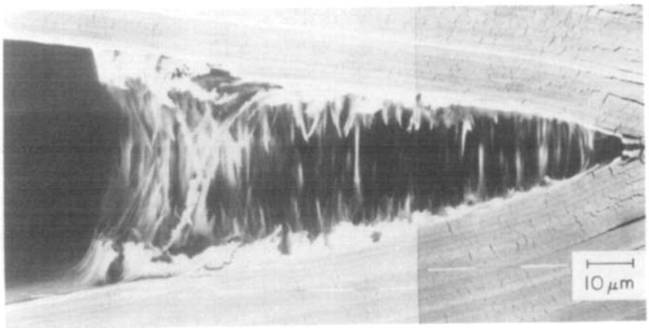

Figure 5 Craze structure in the homopolymer

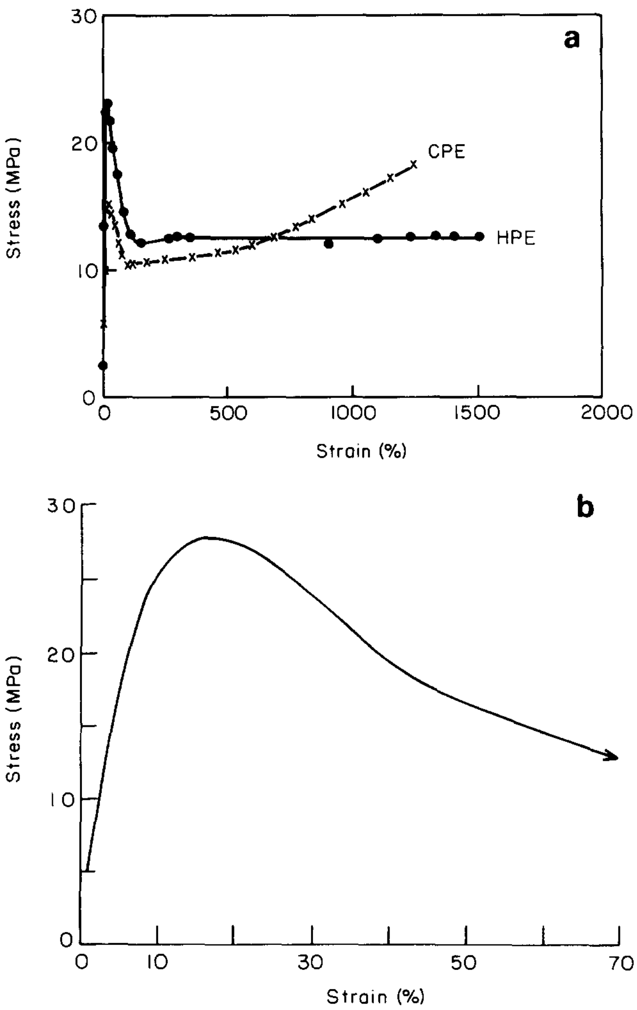

Figure 6 (a) Stress–strain behaviour of the homopolymer and copolymer ($\dot{\varepsilon}=0.4\,\text{min}^{-1}$, $T=42°C$). (b) Stress–strain curve of homopolymer

craze gives an indication of the boundary stress at that point.

It is of interest to derive the stress distribution corresponding to the observed strain distribution. The stress–strain curve for the HPE material (*Figure 6a*) will be used for guidance. First note that the maximum strain at the craze tip is 12%. This value corresponds to the yield strain. Thus, it is suggested that the yield stress probably exists at the craze tip. This suggestion is reasonable because the craze tip is the point at which newly crazed material is produced by a yield process. The decrease in strain beyond the craze tip in the direction of the notch is consistent with the stress–strain curve that exhibits a large decrease in stress after the yield point. The continuous decrease in strain to zero at the notch tip indicates that the fibrils have become very weak and fibril fracture has occurred.

The stress distribution can be determined if it is assumed that the stress on the boundary is related to the observed strain in the same way as the stress is related to the strain in the stress–strain curve. Since the observed strains are less than the yield strain, it is necessary to use the initial part of the stress–strain curve as shown in *Figure 6b*. Thus in *Figure 7* is plotted the stress distribution corresponding to *Figure 4*. These values of the stress probably do not correspond to the absolute values of the stress that existed at the time the photograph in *Figure 3* was taken, because the stress relaxation occurred between the time of the photograph and the loading of the specimen, whereas the stress–strain curve of *Figure 6b* is based on a comparatively short loading time. However, the shape of the stress distribution probably reflects the variations in strength of the craze along the boundary. Probably it is best to normalize the stress relative to the yield point in order to have the best picture of the stress field on the boundary of the damage zone. *Figure 7* shows that the stress far from the craze approaches an equilibrium value of $0.4\sigma_y$, which should correspond to the stress applied to the slice when the notch was opened in the jig.

Figure 8 shows the SEM picture of a copolymer of PE. This specimen prior to slicing had a notch depth of 2 mm and was exposed to a stress of 4 MPa at 80°C for 106 min, which produced a *COD* of 80 μm. After slicing, the *COD* was opened to 103 μm, which is greater than the value under the original loading. *Figure 9* shows the strain distribution corresponding to *Figure 8*. In this case there are two maxima in the strain, one at the craze tip and the other at the notch tip. The strain along the craze is generally about 20%, which is larger than the yield strain (~12%) for the copolymer. This result indicates that plastic strain was introduced when the slice was loaded at room temperature. However, the prominent microstructural features of the craze were produced during the original loading at 80°C.

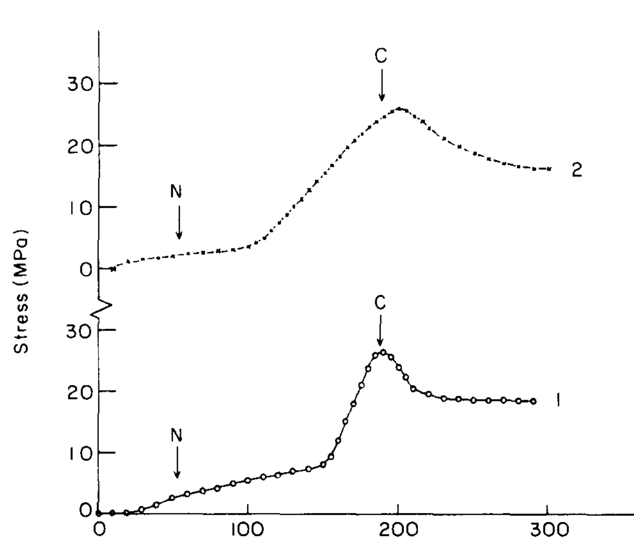

Figure 7 Stress distribution along the damage zone in the homopolymer

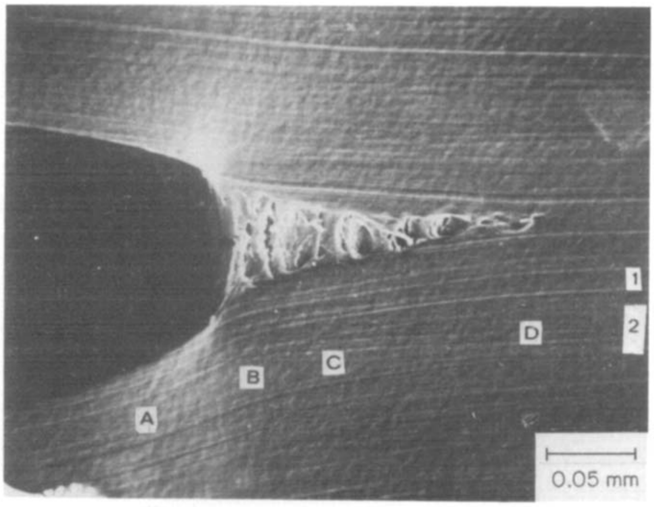

Figure 8 SEM picture of copolymer used for measuring strain field

Figure 8 tells us something about the strength of the fibrils in the copolymer. These results suggest that fibrils near the notch tip are stronger than those further away. This result is consistent with the fact that the copolymer exhibits orientation strengthening (*Figure 6a*).

DISCUSSION

When a plastic strain is measured that is significantly greater than the yield strain, the stress field is not unique. If the specimen is unloaded and then reloaded without producing additional plastic strain, then the difference in strain between the successive loadings should more directly relate to the stress field.

It is important to realize that, between the time the scratched specimen is first loaded and the time the SEM picture is taken, an appreciable amount of stress relaxation will take place. Thus the usual fast stress–strain curve does not give a direct connection between the measured strain field and the relaxed stress field. This technique is newly developed and the transformation from the measured strain to the determination of the absolute value of the stress field will require much additional experimentation. However, knowing the strain distribution along the boundary gives useful information about variations in strength of the fibrils along the damage zone. The experimental method is applicable to all polymers, but the specific results apply to the PE resins used in this investigation.

SUMMARY

A new direct method for measuring the strain distribution in the neighbourhood of a craze produced under plane strain conditions has been developed. Information about the corresponding stress distribution can be obtained directly from the measured stress–strain curve of the polymer.

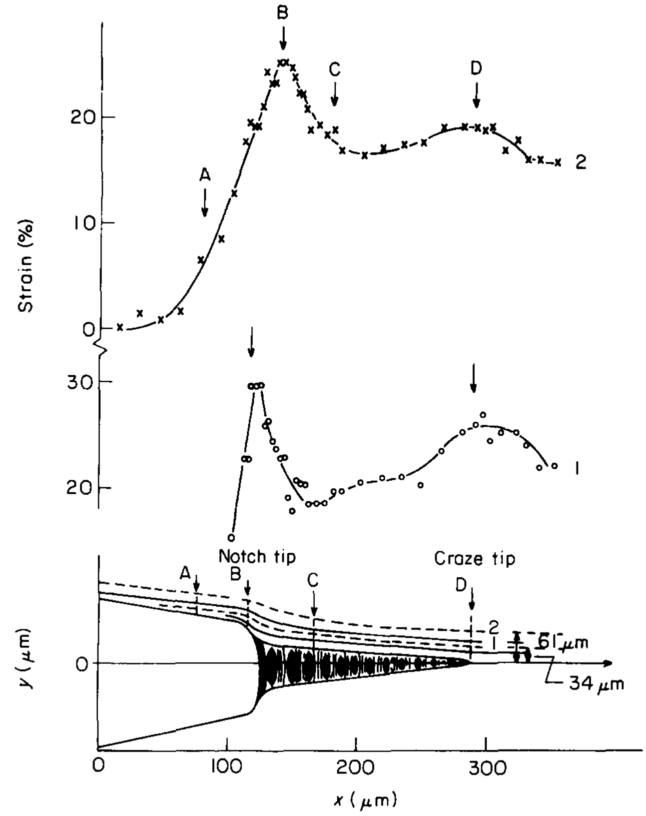

Figure 9 Strain distribution along the damage zone in the copolymer (TR-418. Notch depth =2 mm; σ_1=4 MPa; T=80.0°C; t=106 min; $CMOD$=625 μm, initial COD=86 μm, final COD=103 μm

ACKNOWLEDGEMENTS

The research was sponsored by the US Department of Energy and by the Gas Research Institute. The Central Facilities of the LRSM Laboratory as supported by NSF were very helpful.

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