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# Crack initiation and fibre creep in polyethylene

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A critical appraisal has been carried out of the creep performance of the fibrils in the pre-crack damage zone (craze) of a series of polyethylene homo- and copolymers. It has been shown that by assuming that the fibrils are drawn to the natural draw ratio of the material, a correlation can be established between the creep rate of oriented specimens and the stress crack performance of the material. The existence of such a correlation paves the way to simpler and faster testing routines and to a better understanding of the factors controlling stress crack performance.

**(Keywords: crack initiation; creep; polyethylene)**

*Introduction*

This paper reports a critical appraisal of the factors affecting crack initiation in polyethylene. In particular, it focuses on the processes involved in the development of localized damage leading eventually to crack formation. It identifies a key mechanical parameter which relates to the material crack performance and discusses the scope for performance control through molecular structure.

Fresh interest in this important area has been generated by the systematic work of Lu *et al.*[1]. They have shown that in a sharply notched specimen under static tensile loading, a wedge shaped damage zone is formed at the notch tip, which increases in size at a linear rate until crack growth begins. The rate at which the damage zone develops is orders of magnitude slower for copolymers than for homopolymers[2,3]. Furthermore, it was shown that this zone consists of highly voided material with fibrillar structure.

It was recognized that the initiation mechanism involves two processes, one being the yield process converting essentially isotropic material to the fibrillar form, and the second being the creep of the fibrils up to the point of fibril rupture. The fibril creep was viewed as the dominant mechanism in copolymers.

An extensive programme of work is currently underway in our laboratories to test and develop these hypotheses. As part of this programme, the relevance has been examined of an approach previously applied by Ward and Wilding[4,5] to the study of very highly drawn polyethylene. The preliminary results reported here offer a stimulating new perspective on the relationship between creep and long term performance of polyethylene.

*Experimental*

The programme was based on three materials of widely differing structure and fracture performance, as detailed in *Table 1*. Sheets of about 130 μm thickness were compression moulded at 180°C and crash cooled in the press. In order to simulate fibrils, small dumb-bells 20 mm long by 5 mm wide were stamped from the sheet, and drawn to their natural draw ratio ($\lambda_n$) at 10 mm $min^{-1}$ and 23°C. The draw ratio was defined by the separation of dots marked at 2 mm intervals prior to drawing. Yield stress was measured during this operation, and drawing was halted when the yield neck had propagated to the shoulders of the dumb-bell.

The drawn central portion of each dumb-bell was measured for width and thickness, and fitted to a creep machine of the type described in ref. 5.

The stress to be applied to each specimen was calculated to match the stress actually experienced by the individual fibrils in the damage zone. The following consideration was used. For copolymers, Wang and Brown[6] reported stresses at the notch tip close to the value of the yield stress ($\sigma_y$) for the bulk polymers. Thus, if we assume that the fibrils in the damage zone are drawn to the natural draw ratio of the material, then the stress to be applied in the creep experiment is given by:

$$\sigma_t = \sigma_y \lambda_n$$

The creep machines were maintained at 23 ± 1°C and the extension was recorded automatically for a period of about 24 h or until specimen breakage occurred, if this was sooner.

Stress crack performance of each material was assessed by measuring the lifetime in the bottle stress crack (BSC) test[7]. The time for a crack to grow from a stress concentration in a blow moulded bottle containing a detergent solution was obtained by monitoring automatically the electrical resistance of the bottle wall. The test was terminated when the resistance decreased to a preset level due to crack growth.

**Table 1** Characterization of materials used

| Sample | Density ($kg\ m^{-3}$) | Branch conc. (per $10^3$ C) | $\bar{M}_w$ | Bottle stress crack: Lifetime (h) | Bottle stress crack: Test temp. (°C) |
|---|---|---|---|---|---|
| HP | 960 | 0 | 130000 | 1 | 60 |
| CP1 | 953 | 1.5 | 160000 | 4 | 60 |
| CP2 | 938 | 4.5 | 180000 | >1000 | 80 |

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*Results and discussion*

Values of yield stress, natural draw ratio and applied creep stress are shown in *Table 2.* As branch concentration increases, both yield stress and natural draw ratio decrease. Consequently, the applied stress decreases.

The creep data for the three samples are shown in *Figure 1.* There is a strong parallel with the curves reported previously for the time dependence of damage growth at the notch tip in homo- and copolymers[2,3], suggesting that the two phenomena may indeed have a common origin. Even more striking, however, is the correlation between fibre creep and bottle stress crack performance (cf. *Figure 1* and *Table 1*). The homopolymer HP exhibits a rapid increase in strain with time and has a low value of BSC lifetime. For the copolymer CP2, on the other hand, the rate of creep decreases sharply with time and the strain tends to a finite limiting value. The BSC lifetime for this material is very long, more than three orders of magnitude longer than that of the homopolymer.

**Table 2** Yield stress ($\sigma_y$) and natural draw ratios ($\lambda_n$) of the three materials used, and stress levels applied during the creep experiments ($\sigma_t$)

| Sample | $\sigma_y$ (MPa) | $\lambda_n$ | $\sigma_t$ (MPa) |
|---|---|---|---|
| HP | 26 | 10 | 260 |
| CP1 | 20 | 8 | 160 |
| CP2 | 16 | 6 | 116 |

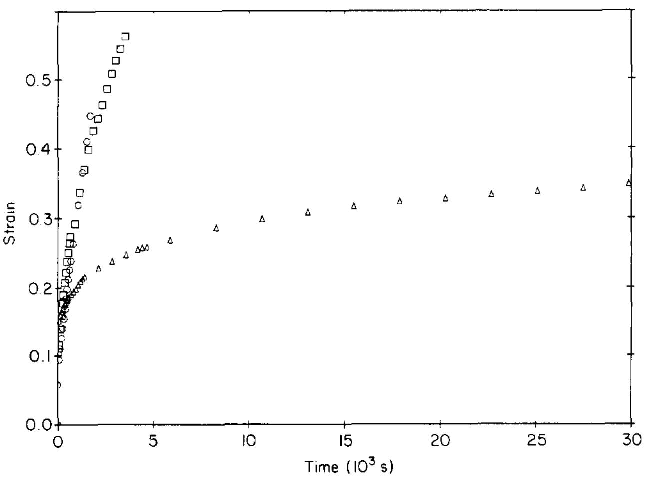


**Figure 1** Room temperature creep data for samples of the three materials drawn to their natural draw ratio. Stress levels are as shown in *Table 2.* ○, HP; □, CP1; △, CP2

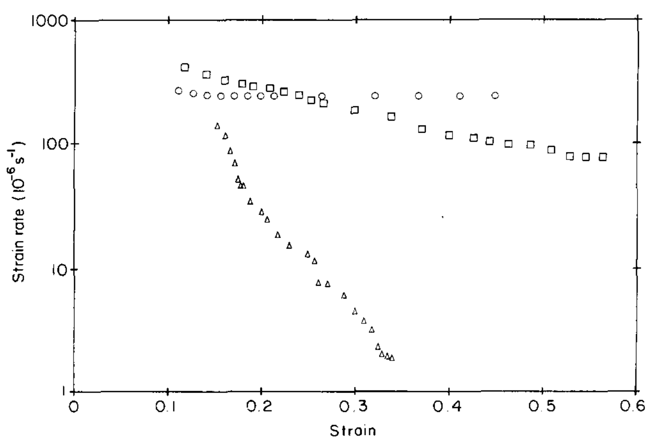


**Figure 2** Sherby–Dorn plots derived from the data in *Figure 1.* ○, HP; □, CP1; △, CP2

The representation of *Figure 1* is not sufficiently refined to discriminate between the homopolymer and the low branch content copolymer CP1. It will be shown below that this limitation can easily be overcome.

Although the message conveyed by the data in *Figure 1* and *Table 1* is clear, its quantitative expression may be ambiguous, because the criterion to express relative creep performance is not defined. This problem is overcome by using a different representation of the creep data, in strain rate *versus* strain plots (Sherby–Dorn plots). This approach has been used successfully by Ward and Wilding[4] in their work on very highly drawn polyethylenes.

Sherby–Dorn plots for the three materials of the present study are shown in *Figure 2.* Two features are immediately apparent. First, this type of representation discriminates effectively between samples of different stress crack resistance. HP settles quickly to a constant plateau strain rate, whilst CP1 exhibits a strain rate decreasing gradually with time, without any indication of a plateau value. CP2, on the other hand, is characterized by a rapid drop in strain rate with increasing strain. Second, the slopes in *Figure 2* provide a sensitive tool for the comparative evaluation of materials with a view to predicting their relative stress crack performance. An obvious advantage of this approach is that the creep experiment requires only a small amount of material. Moreover the analysis of the creep response becomes a powerful basis for the investigation of the molecular process controlling crack resistance.

Finally it should be noted that whilst fibril creep emerges as a major factor influencing stress crack performance, it cannot be the only one. If it were, neither CP1 nor CP2 would show the constant growth rate of the damage zone reported by Brown and co-workers[1,2].

Further drawing of the isotropic material must contribute to the increase in length of the fibrils. Thus the full interpretation of the experimental observations should take into account the interaction between the processes of yielding and fibril creep which proceed simultaneously.

*Conclusions*

In a number of cases a link has been established between creep response and the craze (pre-crack damage) propagation pattern in different types of polyethylene. The relative stress crack performance of a set of homo- and copolymers was successfully predicted using simple tensile creep tests on drawn specimens.

As creep response is sensitive to both molecular orientation and level of applied stress, the appropriate choice of these two parameters is critical to the successful development of this approach. Experimentally determined values of the stress in the crazed zone were used. The fibrils (in the craze) were assumed to be drawn to the natural draw ratio of the material.

Although creep appears to be a major factor in controlling the early stage of damage propagation, the fine details of the process preceding the appearance of a crack are also influenced by yielding and neck propagation.

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