# On the Tensile Behaviour of Oriented Polyethylene

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Tensile tests of oriented, high-density, polyethylene films were carried out in order to assess how far the intrinsic molecular and textural features of the samples affected their mechanical behaviour, and to see whether this behaviour bore any resemblance to that of metals. Brittle fracture, ductile fracture or necking followed by drawing occurred, depending on the pretreatment used to orient the material, the angle between the tensile axis and the *c* axis, and the speed of testing. The feature of the crystalline texture (common to all the films) which was important in determining the geometry of deformation was the alignment of the *c* axis; the additional orientations present in some of the films did not significantly affect the deformation geometry. The results suggested that ductile deformation approximated to slip in the direction of the *c* axis, while brittle fracture was observed to occur on planes parallel to the *c* axis. There were some noticeable differences in behaviour between those films which had received a pre-anneal and those which had not; these differences appeared *not* to be due to differences in the crystalline texture. Several striking similarities to the mechanical behaviour of hexagonal single crystals were observed.

### 1. Introduction

The tensile behaviour of polymeric materials is clearly of great importance for many applied problems, particularly at high deformations in the region of yield and fracture. The corresponding effects in simpler materials, such as metals, are well documented and reasonably well understood both in terms of general plastic behaviour of matter, and in terms of atomic structure. In the case of polymers, the situation is much more obscure as regards both these facets. Our outlook is much influenced by the unique atomistic feature of these materials, and there is a tendency to regard all observations as consequences of the long-chain nature of the molecular constituents. Lately, our microstructural picture has been greatly extended by the manifold morphological features recognised in crystalline polymers, which are now gradually being taken into account by current works on macroscopic properties. On the other hand, a number of macroscopic structural effects have been reported in the course of yield and fracture, which resemble features long familiar in non-polymeric substances [1, 2].

The present work, the commencement of a longer-term research project, has been undertaken in order to assess how far yield and fracture phenomena, occurring on tensile testing, are attributable to molecular and textural features intrinsic to the polymeric nature of the samples, and how far there is a common denominator with simple substances such as metals.

For the purpose outlined, we chose samples which are as well defined as possible, in our present state of knowledge, as regards molecular orientation and crystalline texture. The basis for this is provided by recent work on polyethylene with which one of us was associated [3, 4]. In the course of this, polyethylene was first drawn, then rolled along the draw direction and subsequently heat treated appropriately. This led to structures which were well specified in terms of macroscopic sample dimensions both with respect to molecular structure and crystalline texture. Amongst others, macroscopic samples were obtained where the three crystal axes a, c, cand b were uniquely defined by the rolling-plane normal (a), rolling and drawing direction (c), and by the direction in the film (rolling) plane perpendicular to the draw-roll direction (b). In these samples, the planes of the crystal lamellae had a well-defined double texture with the lamellar planes parallel to b and inclined by about  $40^{\circ}$  with respect to c. This was the basic sample type used in this work, together with three others less uniquely specified in terms of macroscopic dimensions serving as controls (see below).

It ought to be stated at the outset that the influence of the orientation, both on the level of the crystal lattice and that of the crystal lamellae (with the exception of the c axis position itself), was not as decisive as originally anticipated, at least under the test conditions in question, and that there emerged a closer resemblance to metals than is normally recognised. On the other hand, there were some differences which may become important for further work to be discussed in the paper. Having indicated these facts to begin with, the experimental material will be presented in the order in which it was originally obtained.

## 2. Experimental

### 2.1. Samples

The starting material was a film of high-density polyethylene (Marlex 50)  $4.9 \times 10^{-2}$  cm thick. It showed little preferred orientation as judged from wide-angle X-ray diffraction photographs. Preparation of the doubly oriented, basic, sample-type material referred to above was carried out in the light of the work of Hay and Keller [4]. The process consisted of roomtemperature drawing of a strip of the material (draw-ratio about 6, the thickness being reduced by a factor of about 3 and the width perpendicular to the draw direction by a factor of about 2), followed by room-temperature rolling with the direction of advance in the draw direction, followed by annealing at a suitable temperature with the film held between sheets of mica in a silicone oil bath. The best annealing temperature was found by experiment to be 131.1° C. The rolling resulted in a 15% decrease in thickness and a 5% increase in width perpendi-390

cular to the draw direction, with a corresponding 10% increase in length in the draw direction. The annealing caused a 30% increase in thickness and a 10% decrease in width, accompanied by a 40% decrease in length in the draw direction. Wide-angle X-ray diffraction then showed a satisfactory degree of double orientation of the crystalline material, with the b and c axes aligned parallel to the plane of the film (the latter in the draw direction) and the a axis aligned perpendicular to it. It is convenient to denote the mode of preparation of this film by the code D - R - A (drawn - rolled - annealed). The three control films were then prepared: respectively, by drawing at room temperature (D), by drawing and rolling at room temperature (D - R), and by drawing at room temperature and then annealing at  $131.1^{\circ}$  C (D – A). The dimensional changes were similar to those recorded above. The crystalline textures as determined by wide-angle X-ray diffraction were as follows: D had c aligned parallel to the plane of the film with a and b randomly distributed around it; D - R had c aligned parallel to the plane of the film and also (110) and  $(1\overline{1}0)$ aligned parallel to the plane of the film; D - Ahad c aligned parallel to the plane of the film and in addition a small amount of D - R - Atype double orientation. The presence of double orientation in D - A was unintentional. The amount was small compared with the amount in D - R - A; it was therefore considered reasonable to regard D - A as a valid control sample.

### 2.2. Tensile-Testing Arrangements

Parallel-sided tensile-test-pieces were cut from the prepared films with a razor blade. Most testpieces were in the range length 1 to 3 cm, width 1 to 3 mm, thickness 0.1 to 0.2 mm. The angle between the test-piece axis (i.e. the direction along which the sample was pulled) and the *c* axis was measured in a polarising microscope to about  $\pm 1^\circ$ . This angle is denoted by the symbol  $\lambda_0$  before deformation and by  $\lambda$  after deformation.

A conventional tensile-testing machine was used with the crosshead driven at a constant speed. Most tests were carried out at a standard speed of 1.6 cm/min; in a few cases, the speed was reduced to 1/80 of this. The loads involved were about 1 to 2 lb (1 lb = 454 g) and were measured on a Boulton Paul, 250 lb, load cell, the output from which was fed to a constantspeed chart recorder via the associated bridge circuitry. The load-cell displacement at 250 lb was 0.02 in. (1.0 in. = 25.4 mm), and was proportionally smaller at smaller loads. Tests were carried out with the specimen either in air at room temperature or in water cooled with ice to  $4 \pm 2^{\circ}$  C.

#### 2.3. Results

#### 2.3.1. Tensile Tests at Room Temperature and Standard Speed

The behaviour of all four types of prepared film could be classified under three headings:

(a) Drawing The load, after rising to a maximum (upper yield point), fell to a lower value (lower yield point), and thereafter remained constant or rose slowly. Inhomogeneous yielding took place during the load drop, and the region of greatest elongation then propagated along the test-piece at constant or slowly rising load. (b) Ductile fracture The load rose to a maximum and then fell gradually to zero. At some stage during the load drop, the specimen began to break by a tearing process which could be arrested by stopping the tensile-testing machine. (c) *Brittle fracture* The test-piece broke in two after inappreciable plastic extension, the breaking being too quick to be arrested by stopping the tensile-testing machine.

The circumstances in which these types of behaviour occurred are shown in table I. (D – A tested at  $\lambda_0 = 0^\circ$  was unusual in breaking suddenly rather than gradually after appreciable deformation.) As seen, the major difference between the four samples is the increased angular range of brittle fracture in the D – R – A and D – A (i.e. the annealed) materials.

In test-pieces which exhibited drawing, it was observed that with increasing elongation the c axis swung round towards the tensile axis (i.e.  $\lambda$  decreased from its initial value of  $\lambda_0$ ), becoming parallel to it in the most highly extended portions. The ratio of the width of an unextended test-piece to that of the same test-piece when fully extended was very much greater than the corresponding thickness ratio. Width ratios increased with  $\lambda_0$  from about 4 at  $\lambda_0 = 15^\circ$  to about 10 at  $\lambda_0 = 90^\circ$ , whereas thickness ratios were all between 1.1 and 1.3 and appeared not to vary systematically with  $\lambda_0$ .

During the load drop, the deformation became clearly inhomogeneous along the length of the test-piece. In certain circumstances, the inhomogeneity was rather striking: one or more deformation bands were formed, across which there was high local shear (shear angles of tan<sup>-1</sup> 10 were often seen) accompanied by abrupt steps on the edge or edges of the test-piece (bands did not always pass right across the testpiece). A deformation band is illustrated in fig. 1. It appears to be like some of the bands reported by Kurokawa and Ban [1] (see their fig. 1b and fig. 2b, upper diagram). The boundaries of the band were approximately plane, and intersected the wide surface of the test-piece approximately at right-angles. Close examination of several of these bands showed that the wide surface of the test-piece was grooved where the band intersected it. This indicates that the deformation in the band was not just a simple shear parallel to the boundaries of the band. The sum of the depths of the grooves on the two sides was some 5 to 10% of the test-piece thickness. Often the band was observed to be not quite straight, but to have slight curvature in the opposite sense at the two ends. The boundaries appeared to be smooth when seen at magnifications up to 320. When  $\lambda_0$  was less than about 40°, the bands were parallel to the c axis in the adjacent material, within the uncertainty of measurement: for larger values of  $\lambda_0$ , there were increasing devia-

TABLE I Behaviour at room temperature and standard speed as a function of  $\lambda_0$ .

Material	Behaviour					
	Ductile fracture Drawing		Brittle fracture			
	$\overline{\lambda_0}$					
D	<b>0</b> °	15 to 90°	Did not occur			
D – R	<b>0</b> °	15 to 85°	Only occurred at 90°			
D – R – A	0 to $15^{\circ}$	30 to $70^{\circ}$	75 to 90°			
D – A	0* to 15°	30 to $73^{\circ}$	75 to 90°			

\*At  $\lambda_0 = 0^\circ$ , broke suddenly after appreciable deformation.



Figure 1 D – A test-piece,  $\lambda_0 = 60^\circ$ , showing the large shear across a deformation band. Angle between band and c axis is 8° (× 12).

tions, up to a maximum of  $17^{\circ}$ , between the deformation boundary and the direction of the c axis, always in the sense that the acute angle between the deformation boundary and the test-piece axis was less than that between the c axis and the test-piece axis. Denoting the angle between the c axis and the deformation band by  $\alpha$ , then  $\alpha$  varied with  $\lambda_0$  as shown in fig. 2. It will



Figure 2  $\alpha$ , the angle between deformation band and c axis, plotted against  $\lambda_0$ , the initial angle between c axis and tensile axis, for D – R – A and D – A tested at room temperature and for D tested at 4° C.

be observed that the room-temperature data in this figure are confined to D - R - A and D - A. This is because deformation bands of this kind were not found in D and D - R in room-temperature tests, although they were found at 4° C, as described in section 2.3.3. The data go only up to  $\lambda_0 = 73^\circ$  because, above this angle, D - R - A and D - A suffered brittle fracture without developing deformation bands. In the tests in which  $\alpha$  was measured, the testing machine was stopped as soon as it was appreciated that a deformation band had formed. The angle  $\lambda$  between test-piece axis and c axis in the material adjacent to the band was less than its initial value  $\lambda_0$  because of deformation. Although 392

it was not known at precisely what moment the band was formed, it was thought that at the moment of formation this angle was closer in value to  $\lambda$  than to  $\lambda_0$ . For this reason,  $\alpha$  was also plotted against  $\lambda$ , as in fig. 3. The effect has been to reduce the scatter; within experimental error all points for D – R – A and D – A, except two, lie on the same smooth curve, which diverges from  $\alpha = 0^{\circ}$  as  $\lambda$  rises above 20°.



*Figure 3*  $\alpha$ , the angle between deformation band and c axis, plotted against  $\lambda$ , the angle between c axis and tensile axis measured after formation of band, for D - R - A and D - A tested at room temperature and for D tested at 4° C.

All test-pieces which draw with continued extension necked, whether or not deformation bands were formed. In some cases, the transition region between the highly and slightly deformed regions had long gently-tapering sides; in other cases, the change in width occurred abruptly and in a characteristically asymmetrical way, which suggested that shear had taken place in a direction approximately parallel to the *c* axis. An example of the abrupt type of transition region in a D – R test-piece with  $\lambda_0 = 75^\circ$  is shown in fig. 4. The set of fine, parallel lines, which origin-



Figure 4 Shoulders of neck in a D – R test-piece,  $\lambda_0 = 75^{\circ}$ , after drawing in the tensile test. The closely-spaced parallel lines run everywhere parallel to the direction of the *c* axis; a sharp kink boundary is clearly visible ( $\times$  6).

ated naturally in the preparative drawing process and survived the subsequent rolling, are parallel to the *c* axis. (The abrupt change of orientation across a straight boundary, which these lines reveal, is referred to in the next paragraph.) Abrupt transition regions were associated with high values of  $\lambda_0$  and often also with proximity to a grip, either because the neck formed near a grip or because the transition region approached a grip during the test.

As was mentioned above,  $\lambda$  decreased with increasing deformation. Thus  $\lambda$  changed along the length of the transition region. The change was sometimes abrupt across a straight boundary, here named a "kink" boundary. An example of a kink boundary can be seen in fig. 4, running obliquely across the test-piece; the change in orientation across the boundary is revealed by the change in direction of the fine lines which run everywhere parallel to the c axis. It can be seen that the kink boundary did not bisect the angle between the c axes on either side of the boundary; this was typical, the angle between the kink and the bisector ranging from 4 to  $18^{\circ}$ in the cases examined. Kink boundaries were commonly seen in the abrupt transition regions in D - R and D test-pieces, not so often in D - R - A and D - A test-pieces. Kink boundaries were sometimes seen in the neighbourhood of the curved parts of deformation bands. In a few cases, such boundaries were seen at the very start of yielding, before substantial deformation had occurred. They were also seen in the D - Rtest-pieces at  $\lambda_0 = 90^\circ$  which suffered brittle fracture. One such test-piece is pictured in fig. 5.



*Figure 5* D – R test-piece,  $\lambda_0 = 90^\circ$ . Brittle fracture has occurred parallel to the *c* axis. The lenticular region running in from the edge of the test-piece at about 30° is dark because its orientation differs significantly from that of the remainder of the material (× 6).

The kink boundaries enclose the darker lenticular region, darker because of its different orientation, which makes an angle of about 30° with the edge of the test-piece. It appears that the specimen attempted to yield here but could not avoid brittle fracture. It was not necessarily the case that kinks formed early and persisted throughout the test. One ductile test-piece was observed at intervals throughout a test. A kink boundary developed before necking occurred but disappeared as a gently tapered neck was formed. Later, one of the transition regions moved towards a grip, became more abruptly tapered, and a second kink appeared.

The upper and lower nominal yield stresses of the four materials have been plotted against  $\lambda_0$ over the drawing range in figs. 6, 7, 8<sub>2</sub> and 9. The nominal stress is the load divided by the initial area of cross-section of the test-piece. The variation of the yield stresses with  $\lambda_0$  is qualitatively the same in all four materials; the scatter in the case of D - R - A is rather higher than would be expected from the estimated measurement errors. The curves for upper and lower yield stresses are plots of the functions  $A(\sin \lambda_0 \cos \lambda_0 + k \sin^2 k)$  $\lambda_0$ )<sup>-1</sup> and B cosec  $\lambda_0$  respectively, where k, A, and B are constants whose values, given in table II, have been chosen to give the best fit with the experimental points. The reason for using these functions is discussed in section 3; at this point, A and B may be regarded simply as convenient parameters with which to compare the upper and lower yield stresses of the different materials. As regards the room-temperature tests, the materials fall into two groups, the annealed and the unannealed; the former had yield stresses about twice those of the latter. In each group, the rolled had slightly higher yield stresses than the unrolled, but these differences were distinctly smaller than those between the two groups.

There is little to report at this stage of the work about the ductile and brittle fracture behaviour. The former was a tearing process usually initiated at the grips. The test-pieces which suffered brittle fracture broke along planes parallel to the *c* direction and perpendicular to the wide face of the test-piece, after inappreciable plastic extension. An example of such a break in a D – R specimen with  $\lambda_0 = 90^{\circ}$  has already been given in fig. 5. The nominal tensile fracture stresses for brittle fracture are plotted in figs. 7, 8, and 9; the values may not be very reliable, as breaks often occurred near the grips.



Figures 6, 7, 8, and 9 Upper and lower nominal yield stresses and brittle fracture stress at room temperature plotted against  $\lambda_0$ , the initial angle between the tensile axis and the c axis for D (fig. 6), D - R (fig. 7), D - R - A (fig. 8) and D - A (fig. 9). (Fig. 6, top left; 7, top right; 8, bottom left.) 394

Material	Room	Room temperature			4° C		
	k	A (10 <sup>8</sup> dyn/cm <sup>2</sup> )	<i>B</i> (10 <sup>8</sup> dyn/cm <sup>2</sup> )	k	A (10 <sup>8</sup> dyn/cm <sup>2</sup> )	<i>B</i> (10 <sup>8</sup> dyn/cm <sup>2</sup> )	
D	0.45	0.87	0.70	0.45	1.4		
D – R	0.40	1.10	0.80	0.40*	1.5		
D - R - A	0.40	2.31	1.80	0.40*	2.8		
D - A	0.45	2.17	1.50				

TABLE II Values of the parameters k, A, and B in the expressions A (sin $\lambda_0 \cos \lambda_0 + k \sin^2 \lambda_0$ )<sup>-1</sup> and B cosec  $\lambda_0$ , which were used to fit the upper and lower nominal yield-stress data respectively.

\*Assumed value.

## 2.3.2. Tensile Tests at Room Temperature and Slow Speed

A few tests were carried out at a speed 1/80 of standard, in order to find out whether this caused the behaviour to change from brittle to ductile. The change of speed did indeed have this effect on D – R test-pieces with  $\lambda_0 = 90^\circ$  and on D – A test-pieces with  $\lambda_0 = 77^{\circ}$ ; these necked and drew at constant load in the usual ductile manner. However, D - A and D - R - A testpieces with  $\lambda_0 = 90^\circ$  were still brittle, breaking at the same stresses as would have been expected in a standard-speed test. It will be noted that the test-pieces which did become ductile at the slow speed had values of  $\lambda_0$  only slightly greater than those marking the upper end of the ductile orientation ranges at standard speed (90° compared with  $85^{\circ}$  for D - R, and  $77^{\circ}$  compared with  $73^{\circ}$  for D – A).

2.3.3. Tensile Tests at 4°C and Standard Speed It has already been noted above that deformation bands of the kind illustrated in fig. 1 were not observed in D and D - R test-pieces tested at room temperature and standard speed. One test was carried out at room temperature and at 10 times the standard speed, and this did produce a small deformation band in a D test-piece. As testing at this and higher speeds presented some difficulties, it was decided instead to retain the standard testing speed and to see whether deformation bands would be formed at a testing temperature of 4° C. It was found that bands were indeed formed under these conditions in D and D - R test-pieces, and were similar in all respects to those described in section 2.3.2. The D material was examined more extensively than the D - R, and in it bands were seen in the range  $\lambda_0 = 15$  to 60°; the values of  $\alpha$  are plotted against  $\lambda_0$  and  $\lambda$  in figs. 2 and 3 respectively. In the latter plot, the D material is clearly

separated from the D-R-A and D-A materials.

A few tests with D - R - A and D - A testpieces showed that deformation bands also occurred in these materials at 4° C; indeed, at lower values of  $\lambda_0$ , they were more prominent than in room-temperature tests at the same angles. At 4° C, they were observed in D - R - Amaterial down to  $\lambda_0 = 22^\circ$ , whereas at room temperature they could not be observed in D - R - A below  $\lambda_0 = 45^\circ$ .

In the range  $\lambda_0 = 72$  to 90°, which is approximately the range in which D – R – A and D – A were brittle at room temperature, D was not brittle at 4° C, but it did not show bands of the kind described above. Instead, at  $\lambda_0 = 72$  and 80°, the test-pieces yielded by kinking. A photograph of a  $\lambda_0 = 72^\circ$  test-piece is reproduced in fig. 10. The boundaries between dark and light



*Figure 10* D test-piece,  $\lambda_0 = 72^\circ$ , tested at 4° C, showing two, well-defined, parallel, kink boundaries set up at the start of yielding. The rather poorly-defined parallel lines run everywhere parallel to the *c* axis (× 18).

regions are the kink boundaries, and the fine lines are everywhere parallel to the *c* axis and reveal the difference in orientation across the kink boundaries. Several test-pieces with  $\lambda_0 = 90^{\circ}$ 395 formed two intersecting bands – one prominent band running right across the test-piece and a second, short one at approximately  $90^{\circ}$ to it and running from the edge of the test-piece into the prominent band but no further.

The upper yield stress of the D material was measured between  $\lambda_0 = 15$  and 90°; the values could be fitted by the same function used for the room-temperature results for the same material, with the same value of k but a higher value of A, as shown in table II. As regards D – R and D – R – A, measurements were made at only one and two values of  $\lambda_0$  respectively. Assuming the same value of k as at room temperature, values of A were calculated for these two materials also and are given in table II. Again, lowering the testing temperature raised the value of A in each case.

## 3. Discussion

The first point to consider is to what extent the behaviour of the four materials, D, D-R, D-R-A, and D-A, differed. In general, their behaviour was rather similar. However, three points of difference can be distinguished in the room-temperature standard-speed tests.

(a) The occurrence of deformation bands (fig. 1) in D - R - A and D - A materials but not in D and D - R materials.

(b) The magnitudes of the yield stresses in the drawing range: as described in section 2.3.2, these are comparable by means of parameters A and B, whose values are given in table II.

(c) The angular range of occurrence or nonoccurrence of brittle fracture, as shown in table I.

In each case, the distinction is between the two materials which had been annealed on the one hand, and the two which had not on the other. It is interesting to note that the same three points of difference can be observed in the results reported by Kurokawa and Ban [1] for high-density polyethylene. They used two pre-treatments, drawing in boiling water and drawing at  $30^{\circ}$  C. The former differed from the latter in the way that the annealed differed from the unannealed in our experiments.

The differences between annealed and unannealed material can be bridged by altering the testing conditions. Referring to the three points of difference listed in the previous paragraph:

(a) Deformation bands did occur in D and D – R material when the test temperature was lowered to  $4^{\circ}$  C.

(b) By lowering the test temperature, the magni-396 tudes of the upper yield stresses (A in table II) of D and D – R were brought closer to the room-temperature values of D – R – A and D – A.

(c) By lowering the testing speed, the value of the angle  $\lambda_0$  at which behaviour changed from ductile to brittle was raised. It seems likely that raising the testing temperature would have had the same effect, and that at a higher temperature D - R - A and D - A would have been ductile up to  $\lambda_0 = 90^\circ$ , as D was at room temperature. Certainly Kurokawa and Ban found that their polyethylene, which had first been drawn in boiling water and was then tested at  $\lambda_0 = 90^\circ$ , was brittle below  $60^{\circ}$  C and ductile above  $60^{\circ}$  C. To summarise, it is as if, when one has the four materials at one temperature, one is, to a first approximation, dealing with the same material at two different effective temperatures; D - R - Aand D – A being at a common effective temperature which is lower than than of D and D-R(more precisely, D - R is at a slightly lower effective temperature than D).

It seems unlikely that the effect of annealing can be ascribed to those changes it caused in crystalline texture which were observed by wideangle X-ray diffraction, in view of the marked similarity in behaviour between D - R - A and D – A despite a considerable difference between the amounts of doubly oriented material in them. However, the annealing also caused a contraction in the c direction of about 40% both in D - R - A and D - A material. This contraction was presumably accompanied by structural changes which did not show in the X-ray pictures, e.g. increase in crystallinity, increase in crystal size, molecular refolding. It is suggested that these, rather than crystalline orientation effects, were the factors which were important in determining the effect of annealing on the mechanical behaviour.

On the other hand, the similarities in the general behaviour of the four materials seem likely to have been due to the crystalline orientation feature which all had in common, namely, the alignment of the c axis. Specific observations which indicate the importance of the c alignment are:

(a) With increasing extension, the c axis swung round towards the tensile axis, whether or not kink boundaries were formed.

(b) The change in width of test-pieces which drew down in tensile tests was much greater than the change in thickness.

(c) The direction of deformation bands varied

in a regular way with the direction of the c axis. (d) The asymmetry of abrupt transition regions in necked test-pieces was always related in the same sense to the direction of the c axis (fig. 4). (e) The upper and lower yield stresses varied in a regular way with  $\lambda_0$ .

(f) The planes of brittle fracture were parallel to c.

Deformation features qualitatively similar to the six just listed have all been observed in single crystals of hexagonal metals, as also have kink boundaries and a ductile-brittle transition. These similarities suggest, as a hypothesis, that, when the oriented polyethylene was ductile, the crystalline parts were deforming by shear in the direction of the c axis and on the plane perpendicular to the plane of the film and containing the c axis. (This hypothesis was also put forward by Kurokawa and Ban [1].) This is certainly an approximation because, as already described, the deformation bands were not in all cases exactly parallel to c, and because the thickness changes in the deformation bands cannot be ignored. If the hypothesis is examined more closely, discrepancies appear:

(a) The deformation bands were not always parallel to c, but showed a monotonically increasing divergence from c with increasing  $\lambda_0$ .

(b) The kink boundaries did not bisect the angle between the directions of the *c* axis on either side. (c) The yield stress results were not strictly in conformity with a critical resolved shear stress criterion for slip on the plane and in the direction specified above. If such a criterion is obeyed for both the onset and the continuation of deformation, and if this slip is the only mode of deformation, the relationship between the nominal tensile stress  $\sigma_{\rm T}$  and the length of the specimen *l*, which arises from the change of orientation brought about by slip is:

$$\sigma_{\rm T} = \sigma_{\rm C} \operatorname{cosec} \lambda_0 \left( 1 - \frac{l_0^2 \sin^2 \lambda_0}{l^2} \right)^{-\frac{1}{2}}$$

where  $\sigma_c$  is the critical value of the resolved shear stress on the slip plane (assumed perpendicular to the plane of the test-piece) and in the slip direction (assumed parallel to the *c* axis). This expression ignores any change of length before yielding. The nominal upper and lower yield stresses  $\sigma_u$  and  $\sigma_L$  are given by

$$\sigma_{\rm U} = \sigma_{\rm C} \, (\sin \, \lambda_0 \cos \, \lambda_0)^{-1} \tag{1}$$

$$\sigma_{\rm L} = \sigma_{\rm C} \operatorname{cosec} \lambda_0 \tag{2}$$

where  $\sigma_{I_i}$  is obtained by putting  $l = \infty$ .

In fact, the upper yield stress data were fitted, as mentioned in section 2.3.1, by the expression

$$\sigma_{\rm U} = A(\sin \lambda_0 \cos \lambda_0 + k \sin^2 \lambda_0)^{-1}$$

rather than by equation 1. This we can interpret by supposing that the resolved normal stress was also involved in the yield criterion: that is, by postulating that yield occurred when the sum of the resolved shear stress and k times the resolved normal stress reached a critical value A. As regards the lower yield stress, the data were well fitted by equation 2. There is a discrepancy here, too, however, for on the basis of equations 1 and 2 we should have  $A = B = \sigma_c$ , whereas in fact B was found to be somewhat smaller than A in every case.

In metal single crystals, the critical value of the resolved shear stress is regarded as independent of normal stress. The introduction of a normal stress term in the present case is clearly a formal necessity in order to account for yielding when  $\lambda_0 \simeq 90^\circ$ . Its significance, however, may well be more than a formal one, as the same constant in the term ensures a fit along the curves to comparatively low values of  $\lambda_0$ . Consequently, the normal stress term may be a genuine physical characteristic. An evaluation, however, would require a more detailed mapping of the deformation geometry (taking into account thickness changes, the exact orientation of the deformation band, etc.) and must accordingly be deferred.

As regards the brittle fracture stress, the results suggest that this might obey a critical, normal, stress law, though the observations were made over too narrow a range of  $\lambda_0$  to be sure of this. The change from ductile to brittle behaviour with orientation at a given test temperature and test speed would then take place at the value of  $\lambda_0$  at which both criteria were reached with the same tensile stress. The expected effect on this value of  $\lambda_0$  of altering the test speed would be the same as was actually observed, namely, that a reduction in speed raised the angle of the ductile-brittle transition.

#### 4. Conclusions

The results provide evidence that the ductile deformation of oriented polyethylene in the conditions described approximated to slip in the c direction within the crystalline regions. The general resemblance to hexagonal, metal, single crystals is remarkable in spite of certain notable differences. Not only is the crystalline part of

the polyethylene a more complex organisation than a metal crystal, there is the additional factor of the presence of uncharacterised amorphous material in the polyethylene. Some of the peculiarities noted in the present work, in particular the deviation of the deformation bands from c, may perhaps be due to such factors, notably to the presence of amorphous, rubberlike material. Nevertheless, it is important to recognise how much of the observed behaviour, particularly under conditions normally encountered in every-day applications, is similar to that of the familiar, simple substances, before specific deductions based on the specifically polymeric nature of the material are attempted.

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