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The plastic deformation of oriented polypropylene: tensile and compressive yield criteria

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Polypropylene, oriented by hot-drawing, has been deformed in tension and compression, at various angles to the initial draw direction. The behaviour in tension is described well by a three-part criterion of the type applied in previous work to fibre-composites [8, 9]. The Hill-von Mises treatment of the plasticity of an anisotropic metal, which has been previously extended successfully to polymers [1], can be applied to describe the tensile yield stresses but is unsatisfactory in its prediction of the form of the strain in polypropylene. In compression, the variation of the yield stress with the angle of the stress axis is much less than in tension, and the modes of deformation are different from those operating in tension at the same angle. In particular, compressing at right angles to the molecules produces shear in a direction normal to the molecules (transverse shear). The critical stress for transverse shear in compression is approximately the same as the critical stress for shear parallel to the molecules in tension.

1. Introduction

The development of criteria for the plastic yielding of polymers which can be applied to a reasonably extensive range of stress conditions has been reviewed recently by Ward [1]. In the case of oriented crystalline polymers, the effect of applying tensile, compressive or shear stress at different angles to the direction of molecular alignment has been studied by a number of workers. In polyethylene terephthalate (PET) [2, 3] and polyvinyl chloride (PVC) [4] the tensile yield stress decreased continuously as the angle between the tensile axis and the molecular direction increased from 0° to 90°. A yield criterion which had been developed by Hill [5] to deal with anisotropic metals (the Hill-von Mises criterion), was used. This satisfactorily predicted not only the orientation dependence of the yield stress, but also the angles at which deformation bands formed in thin sheets. Shear tests on PET however, showed an orientation dependence which could only be accounted for by incorporating an extra term in the Hill-von Mises criterion, representing the effect of an internal compressive stress, or Bauschinger stress [2], tending to contract the extended molecules. The tensile yield stress for polyethylene (PE) showed an upturn as the angle between the tensile axis and the molecular direction approached 90° [6, 7], which was accounted for by a Coulomb criterion, $\sigma_s + k\sigma_n = \sigma_c$ where σ_s is the shear stress in the molecular direction, σ_n the tensile stress normal to the molecular direction and k and σ_c are constants. This criterion, in contrast to the Hill-von Mises criterion, predicts a dependence of the yield stress on hydrostatic pressure. Homogeneous deformation in oriented high density PE was predominantly simple shear parallel to the molecules [6], which supports the use of the Coulomb criterion.

In this paper we report on the behaviour of oriented polypropylene (PP) in tension and compression. It will be shown that the deformation modes in tension are different from those in compression at the same orientation and that the deformation or fracture occurring at different angles of stressing in tension can best be described by a criterion due to Kelly and Davies [8, 9], which was developed to describe the behaviour of composite materials.

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2. Experimental

Tensile specimens were cut from commercial polypropylene sheet*, oriented by hot-drawing. Drawing temperatures in the range 105 to 120 °C, obtained by either an air oven or a silicone oil bath, were used. The initial sheet was 13 mm thick and 57 mm wide, allowing tensile specimens of 3 mm square cross-section to be cut out at different angles θ to the initial draw direction after an elongation of approximately 640%. These were oriented such that the molecular direction lay parallel to one pair of flat faces, the specimens being waisted in this plane (Fig. 4).

Tensile tests were performed on an Instron machine, using "V-block" grips, at room temperature (in the range 22 to 24° C).

Similar specimens were tested in compression between dies of the form shown in Fig. 1. In this case the angle θ is the angle between the compression axis and the molecular direction. In the compressive tests, the initial strain-rate was 1.1×10^{-3} sec^{-1}, and in the tensile tests 1.3×10^{-3} sec^{-1}.

3. Results

3.1. Microstructure

The hot-drawn PP sheet had a fibrillar structure. The drawing was accompanied by a reduction of density from 0.905 to 0.850 g cm^{-3}. The presence of elongated voids or fibrils parallel to the molecular axis was also shown by a pronounced equatorial streak in the low-angle X-ray diffraction pattern.

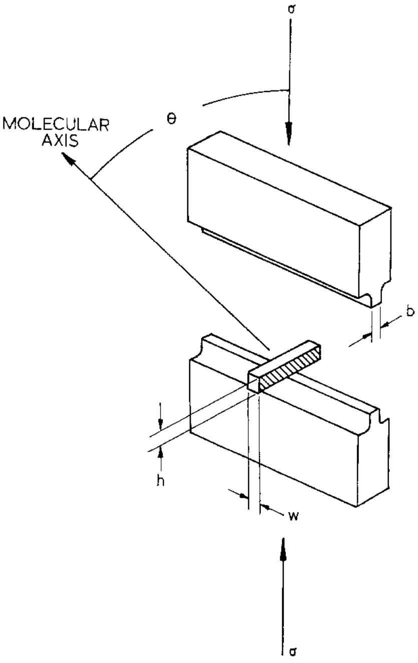

Figure 1 Compression dies and specimen. The dimensions *w*, *h* and *b* are all 3 mm.

Meridional maxima in the small-angle pattern indicated the presence of crystalline lamellae, normal to the draw direction. The wide-angle X-ray pattern was that of the monoclinic form of PP. There was a strong orientation of the molecular direction but no difference in pattern could be detected for different beam directions normal to the draw direction. The material was therefore assumed to have fibre symmetry.

3.2. Tensile tests

A typical set of stress-strain curves is shown in Fig. 2. The variations in drawing procedure described above did not produce significant differences between different sets of specimens. Specimens for which the angle θ between the tensile axis and the molecular direction lay in the range 0° to 10° fractured in a brittle manner, with a jagged, fibrous fracture surface. To prevent these specimens from shearing within the grips, it was necessary to reduce their thickness in the Oz direction (Fig. 4) to about 1 mm. The same was true for $\theta = 20°$ although this specimen did not fracture in a brittle manner. Specimens with values of θ in the range 20° to 80° inclusive, gave large plastic deformations. The deformation was fairly homogeneous in the range 20° to 45°; at higher values of θ, deformation bands of increasing sharpness appeared. An example is shown in Fig. 3. These bands appeared to be similar to, although more diffuse than, bands formed in oriented PE [7] or PET [2]. Although their angle could not be measured with any precision, they appeared to lie near the molecular direction and within the acute angle between the tensile axis and the molecular direction, in agreement with measurements on the more precisely defined bands in PE and PET [2, 7]. These bands were formed in PE and PET specimens cut from thin sheets; the fact that the bands in PP appear in specimens of 3 mm square cross-section is of some significance, as will be shown in the Discussion section below. At higher strains, the bands developed into long necked-down regions.

At $\theta = 85°$ and 90°, ductility was limited by parting in a direction normal to the molecules. The parting was initiated in numerous bands

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*"Trovidur", manufactured by Dynamit Nobel, West Germany.

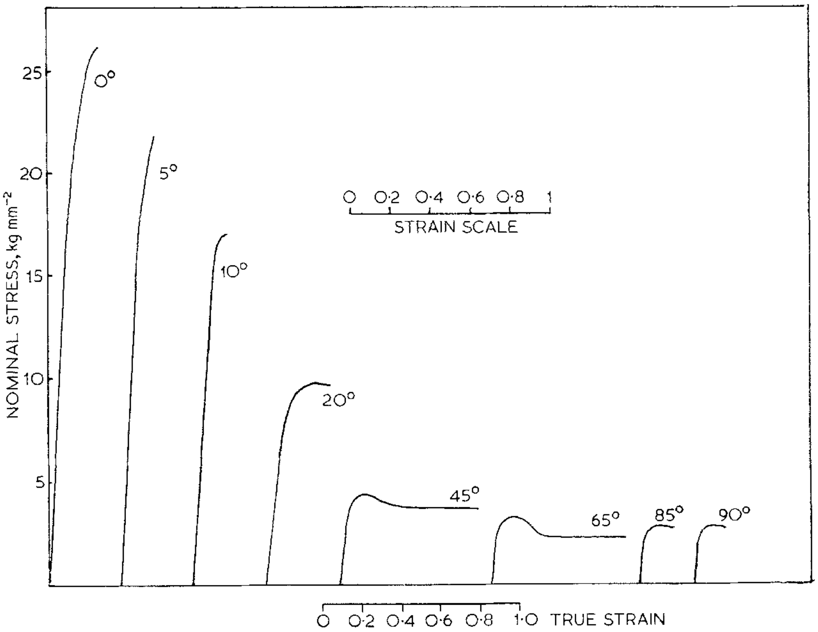

Figure 2 Nominal stress-true strain curves for oriented PP pulled at various angles (marked on the curves) to the molecular direction.

which appeared to be similar to the tensile crazes reported in oriented PET [10], and the final fracture developed from one of these bands. The fracture took place under a gradually decreasing load as the separating fracture surfaces remained connected by a decreasing number of bridging fibrils.

In ductile specimens, the lateral plastic strains ϵ_y' and ϵ_z' were measured (Fig. 4). The strain normal to the molecules ϵ_z' was always much smaller than the lateral strain ϵ_y'. At values of θ in the range 20° to 60° particularly the deformation was to a good approximation a plane strain in the plane defined by the tensile axis and the molecular direction.

3.3. Compression tests

A typical set of stress-strain curves is shown in Fig. 5. All specimens deformed plastically to large strains. It had been thought likely, from the results of the tensile tests, that the strain would occur mainly in the plane containing the molecular direction and the compression axis, especially since the specimen-die geometry imposes a constraint on strain normal to this plane (Fig. 1). However, in all cases a substantial bulging normal to the molecular plane was observed. The magnitude of this increased from the die edges to the die centre, but in all specimens other than $\theta = 90°$, the maximum strain parallel to the dies (i.e. normal to the molecules plane) was of similar magnitude to the strain normal to the dies. At $\theta = 90°$, all lateral strain occurred parallel to the dies, i.e. there was no lengthening in the molecular direction. The effect of the specimen-die constraint could in this case be eliminated by compressing a specimen with the molecular direction parallel to the die. The yield stress was then found to be reduced significantly to approximately 5.0 kg mm^{-2}, and the slope of the stress-strain curve in the plastic region was reduced (Fig. 5). (The yield stress was interpreted as the intersection of the extrapolated elastic and plastic regions of the stress-strain curve.) A further test at $\theta = 90°$ was performed, with the compression axis normal to the larger face of the original drawn sheet, for comparison with the case where the compression axis was parallel to it. The yield stresses for these two directions of compression differed by only 4%, confirming the assumption of isotropy in the plane normal to the molecular direction.

4. Discussion

4.1. Tensile tests

The definition of a yield point is necessarily

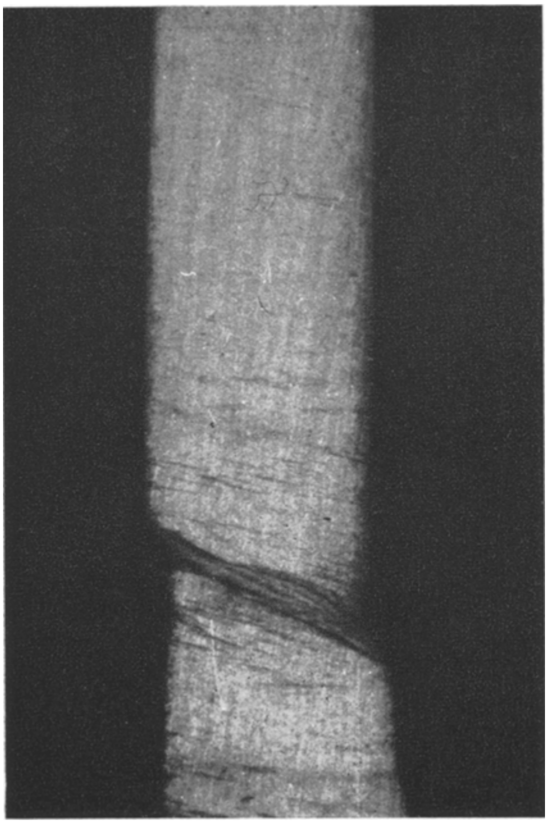

Figure 3 Deformation band in a specimen pulled at an angle of 80° to the molecular direction.

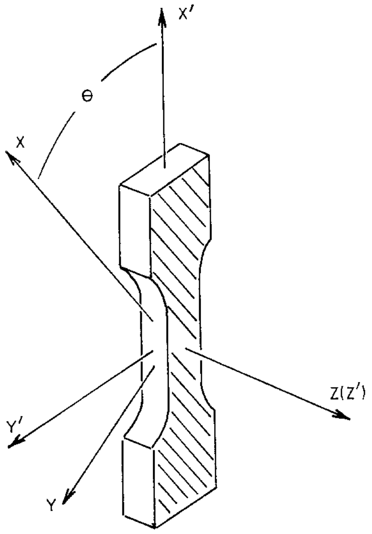

Figure 4 Axes of reference for a tensile specimen. The Ox axis lies in the molecular direction and Ox′ is the tensile axis. Oz and Oz′ are coincident, normal to the plane containing the molecular direction and the tensile axis.

somewhat arbitrary. For specimens which show a load maximum, i.e. for $\theta > 10°$, this may conveniently be taken as the yield point. Specimens of $\theta < 10°$, although relatively brittle, do show a very marked reduction in the slope of the stress-strain curve before fracture. If the knee of the stress-strain curve for these cases is taken to define a yield stress, then the yield stress continuously decreases with θ in the range 0° to 90°, as in PET or PVC [2-4]. A Hill-von Mises yield criterion may then be applied as shown in Fig. 6. The fit at small angles is not good*, but it may be argued that in this region a less well-defined point is taken to describe the yield stress than at higher angles. However, if we turn to the prediction of the form of the strain, the Hill-von Mises treatment for anisotropic metals is found to be inapplicable to oriented PP *even in the range of fully ductile behaviour.* In the Hill-von Mises theory, the plastic strain increment $d\epsilon_{ij}$ is assumed to be given by

$$d\epsilon_{ij} = \frac{\partial f}{\partial \sigma_{ij}} d\lambda \qquad (1)$$

where $d\lambda$ is an arbitrary constant, σ_{ij} is the stress component and f is the yield criterion function

$$f = F(\sigma_y - \sigma_z)^2 + G(\sigma_z - \sigma_x)^2 + H(\sigma_x - \sigma_y)^2 + 2L\tau_{yz}^2 + 2M\tau_{zx}^2 + 2N\tau_{xy}^2 \qquad (2)$$

The axes of reference are the principal axes of the anisotropy and F, G, H, L, M and N are constants. The yield criterion states that yielding occurs when $f = 1$. In our case we take Ox to lie in the molecular direction and Oy to lie in the plane containing the molecular direction and the tensile axis (Fig. 4). Then we have, for an applied tensile stress σ

$$\begin{aligned} \sigma_x &= \sigma \cos^2 \theta \\ \sigma_y &= \sigma \sin^2 \theta \\ \tau_{xy} &= -\sigma \sin \theta \cos \theta \\ \tau_{yz} &= \tau_{zx} = \sigma_z = 0 \end{aligned} \qquad (3)$$

The function f can then be written

$$f = \sigma^2\{(G + H)\cos^4 \theta + (H + F)\sin^4 \theta + 2(N - H)\sin^2 \theta \cos^2 \theta\} \qquad (4)$$

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*In previous papers [2-4], the Hill-von Mises curve is incorrectly shown as having a large negative slope, instead of zero slope, at $\theta = 0$. This makes a significant difference in the range $0° < \theta < 10°$ where however reproducible values of the yield stress are particularly difficult to obtain.

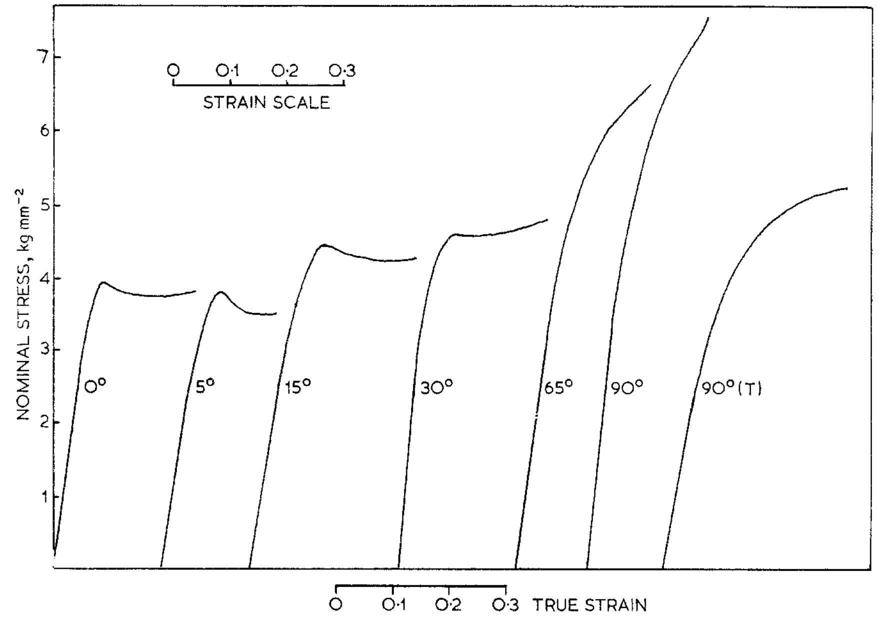

Figure 5 Nominal stress versus true strain for oriented PP compressed at various angles (marked on the curves) to the molecular direction. The specimen 90° (T) had the molecular direction parallel to the long axis of the dies; for all other specimens the molecular direction lay in the plane which was normal to the long axis of the dies (Fig. 1).

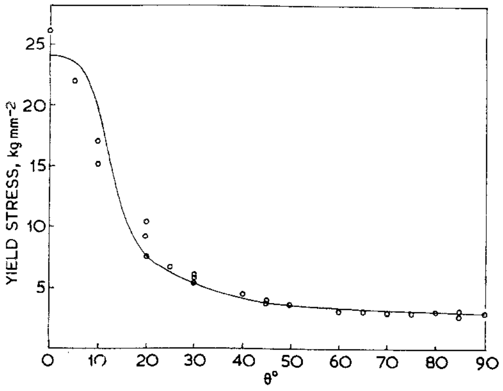

Figure 6 Tensile yield stresses of oriented PP as a function of the angle between the stress axis and the molecular direction. The curve fitted is an equation of the form of the Hill-von Mises yield criterion

$$\sigma = (1.8 \times 10^{-3}\cos^4\theta + 1.11 \times 10^{-1}\sin^4\theta + 1.63 \times 10^{-1}\sin^{-2}\theta\cos^2\theta)^{-\frac{1}{2}}.$$

By fitting the equation $f = 1$ to the yield stress data (Fig. 6)

$$\begin{aligned} G + H &= 1.80 \times 10^{-3} \\ H + F &= 1.11 \times 10^{-1} \qquad (5) \\ N - H &= 8.15 \times 10^{-2} \end{aligned}$$

If we assume transverse isotropy, which is supported by the X-ray diffraction patterns and by the equality, to within 4 %, of the yield stresses for compression in the Oy and Oz directions,

$$G = H \qquad (6)$$

Values of the individual constants can then be determined and the values of $d\epsilon_{ij}$ derived from Equation 1 compared with the experimental observation of a plastic strain confined, to a good approximation, to the Oxy plane.

From Equations 1 and 2

$$d\epsilon_z = -d\eta(F\sin^2\theta + G\cos^2\theta) \qquad (7)$$

where $d\eta$ is an arbitrary constant while the strain increment $d\epsilon_y$ is given by

$$d\epsilon_y = d\eta\{F\sin^2\theta + H(\sin^2\theta - \cos^2\theta)\} \quad (8)$$

The measured lateral strain $d\epsilon_y'$ (Fig. 4) is given by

$$d\epsilon_y' = d\epsilon_x\sin^2\theta + d\epsilon_y\cos^2\theta + 2d\epsilon_{xy}\sin\theta\cos\theta \qquad (9)$$

whence

$$\frac{d\epsilon_y'}{d\epsilon_z'} = \frac{H + (2N - F - G - 4H)\sin^2\theta\cos^2\theta}{F\sin^2\theta + G\cos^2\theta} \qquad (10)$$

Fig. 7 shows the ratio of the lateral strains given by the values of F, G, H and N from Equations 4 and 5. So far from predicting a plane strain,

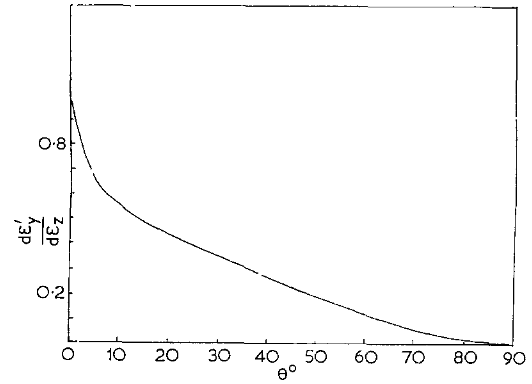

Figure 7 The ratio of the lateral plastic strain increments predicted by the Hill-von Mises theory.

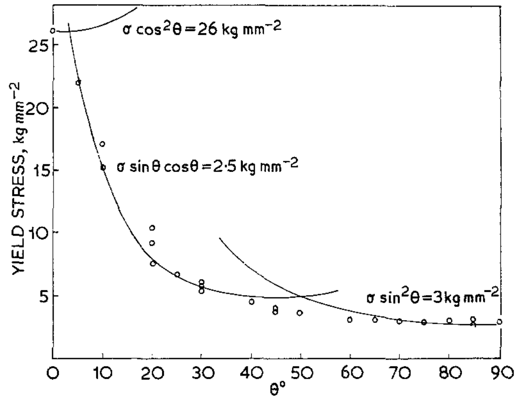

Figure 8 The application of the Kelly-Davies yield criterion to oriented PP.

Equation 10 actually predicts a larger lateral contraction in the Oz direction than in the Oy′ direction. The same deficiency can be seen in the initial formation of deformation bands at higher values of θ. Such bands could not be expected to form in *thick* specimens, against the constraint of undeforming material on either side, if there was an appreciable component of strain in the Oz direction within the band. We must conclude that the Hill-von Mises treatment of anisotropic metals cannot usefully be applied to oriented PP.

A criterion which appears to correspond well with the behaviour of oriented PP is the Kelly-Davies criterion for the maximum stress supported by a fibre composite [8, 9]. This is a three-part criterion:

(*a*) at small values of θ the fibres fail under the component of tensile stress parallel to the fibre axis giving the criterion

$$\sigma \cos^2 \theta = \sigma_c \qquad (11)$$

where σ_c is the tensile strength of the fibres.

(*b*) at intermediate angles failure occurs by shear of the matrix parallel to the fibres, giving

$$\sigma \sin \theta \cos \theta = \tau_L \qquad (12)$$

where τ_L is the shear yield stress of the matrix.

(*c*) near $\theta = 90°$ tensile failure of the fibre matrix interface occurs at a stress σ_u, giving

$$\sigma \sin^2 \theta = \sigma_u \qquad (13)$$

The re-interpretation of these processes as molecular fracture, intermolecular or interfibrillar shear and molecular or fibrillar separation is obvious. The application of the criterion is shown in Fig. 8. Criterion (*b*) could clearly be replaced by a Coulomb criterion which would shift the cross-over point between (*b*) and (*c*) to a higher angle. Otherwise the difficulty that at values of θ up to and including 80°, deformation bands containing large shear components form, can be overcome by supposing that at high angles, such a band is initiated by a molecular parting process, producing regions which then deform in shear. An advantage of the Kelly-Davies criterion is that it yields parameters of immediate physical significance. Thus from Fig. 8, separation of the molecules occurs at a tensile stress of 3.0 kg mm^{-2} and intermolecular shear at a critical resolved shear stress of 2.5 kg mm^{-2}. Detailed confirmation that the deformation mechanisms operating $\theta = 30°$ is a simple shear parallel to the molecules will be given in a later publication.

4.2. Compression tests

The anisotropy of the compressive yield stress is much less than that of the tensile yield stress (Fig. 5), showing that the Hill-von Mises criterion fitted to the tensile data does not apply to the compressive yield stresses. The fact that the compressive yield stress is much lower than the tensile yield stress at small values of θ could be accounted for by the existence of a Bauschinger stress tending to produce contraction in the molecular direction. However, this stress would have to be as high as 11 kg mm^{-2}, and the existence of such a stress seems doubtful in material drawn at 1 kg mm^{-2} at a high temperature, at which internal stresses would be expected to relax quickly. It is more probable that the low compressive yield stress is a result of local buckling activated by intermolecular or interfibrillar shear. This mode of deformation has been observed in oriented PP which was sheared in such a sense that a compressive stress acts in the molecular direction, where it led to the formation of kink bands [11]. No kink bands occurred in our specimens perhaps because the buckling processes were not co-operative over a sufficiently large region. The yield stress for this mode of deformation is difficult to predict but the occurrence of load drops is consistent with a process in which the molecules become progressively more favourably oriented for intermolecular or interfibrillar shear. The mode of deformation at $\theta = 90°$ is more clearly defined. The absence of any extension in the molecular direction shows that the deformation can be accomplished by shear on planes parallel to the molecules in a direction normal to the molecules (transverse shear). Assuming that this shear occurs on planes of maximum shear stress, the critical resolved shear stress for transverse shear derived from the unconstrained compressive yield stress of 5.0 kg mm^{-2} is approximately 2.5 kg mm^{-2}. The operation of two basic modes of deformation, transverse shear and longitudinal shear (shear parallel to the molecules), at similar values of resolved shear stress, together with buckling at small values of θ, accounts for the low anisotropy of the compressive yield stress. The change in shape of the stress-strain curves (Fig. 5) may be accounted for by the rotation of the molecular direction accompanying the longitudinal shear mode, which increases the resolved shear stress for this mode when $\theta < 45°$ but decreases it when $\theta > 45°$. Load drops therefore occur when the initial value of $\theta < 45°$. When the initial value of $\theta > 45°$, the resolved shear stress favouring the transverse component of shear increases as the operation of the longitudinal mode rotates the molecular direction away from the compression axis. However, the operation of the transverse mode is hindered by plastic constraint, leading to a continuously increasing flow stress when $\theta > 45°$.

The Kelly-Davies yield criterion can be applied to the compression results at higher values of θ in the form covering the case where failure of the composite occurs under stress normal to the fibres by shear flow in the matrix. We replace the critical stress for tensile separation of the molecules, σ_u in Equation 13 by the critical stress $2\tau_\text{T}$ where τ_T is the transverse shear yield stress. The two-part criterion

$$\begin{aligned} \sigma \sin\theta \cos\theta &= 2.5 \text{ kg mm}^{-2} \\ \sigma \sin^2\theta &= 5.0 \text{ kg mm}^{-2} \end{aligned} \qquad (14)$$

gives an almost constant yield stress in the range 30° to 90°, in reasonable agreement with the experimental results, within this range. Alternatively, it may be assumed that shear can occur in any direction on planes containing the molecular direction, at the same resolved shear stress, leading to deformation by shear on planes at 45° to the compression axis at the same compressive stress within the range $45° < \theta < 90°$. In this case the shear direction would change continuously with θ, from being parallel to molecules at $\theta = 45°$ to being normal to the molecules at $\theta = 90°$. Further studies of the form of the strain in constraint-free compression tests are needed to determine the most appropriate yield criterion for compression. The present observation that the non-uniform, constrained expansion normal to the plane containing the molecules has a maximum value comparable to the expansion in the molecular plane, for a wide range of initial values of θ, does suggest that a criterion predicting a mixed shear mode may be most appropriate for the compression case.

4.3. General

One of the more surprising results of the present work is that in hot-drawn PP, the critical shear stress for longitudinal shear (parallel to the molecules) in tension is approximately equal to that for transverse shear (normal to the molecules) in compression. Transverse shear within chain-folded lamellae can only occur by breaking or unfolding molecules, except when it occurs on planes containing the chain-folds. Longitudinal shear however can occur without changing the nature of the chain folds. A study by X-ray diffraction of the longitudinal shear process in PP, to be published later, has proved that the main component of longitudinal shear is a homogeneous shear within the crystalline lamellae. The transverse shear mode in compression remains to be studied in detail and it is possible that it takes place between fibrils rather than homogeneously within the crystallites. The transverse shear mode is not observed in tension, where strain is largely confined to the plane containing the molecules and the tensile axis, and the lack of this component of strain appears, essentially, to be the reason for the breakdown of the Hill-von Mises prediction of the form of the strain. It may be a general result that transverse shear occurs only under compression; for example in oriented PE [7], tension at $\theta \sim 90°$ produces either brittle fracture, as in PP, or kinking activated by longitudinal shear.

The general implication of this discussion is that the plastic behaviour of oriented crystalline polymers may require different yield criteria for different types of stress state, and that completely general treatments, such as the Hill-von Mises, may not have a wide range of applicability.

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