

Some Studies of Deformation Processes in Fully-Spherulitic Polypropylene

J. L. WAY, J. R. ATKINSON

Department of Metallurgy, The University of Leeds, Yorks, UK

The deformation of isotactic polypropylene has been studied by optical microscopy of thin sections cut from bulk tensile specimens. An attempt has been made to relate the three different deformation zones within a spherulite to the cross-hatched structure peculiar to polypropylene.

1. Introduction

The deformation of semicrystalline polymers is usually considered in terms of a spherulitic structure, though study has also been made of structures with a relatively large, amorphous, interspherulitic region [1, 2]. Practical considerations have usually resulted in work being carried out on thin films, either solution cast [3] or more frequently melt cast [4], but this leaves unanswered the question of whether the deformation noted can be applied to bulk samples. There is evidence that, in these thin films, interspherulitic cracking occurs due to a thickening of the centres of the spherulites at the expense of the edges, an effect which obviously has a profound effect on the deformation characteristics of these films. There is also the suggestion [2] that, for polypropylene at least, there is a structural difference between these essentially two-dimensional spherulites and the three-dimensional bulk spherulites. Hence it is important to attempt to study the deformation of bulk specimens and thereby avoid spurious thickness effects.

Most work on the deformation of semicrystalline polymers has been confined to polyethylene, for this has many advantages in handling and in ease of interpretation with respect to its relatively simple molecular structure. It is usually considered as representative of many spherulite-forming polymers. However, each polymer has its own characteristics and in this paper we are concerned with the cross-hatched structure peculiar to isotactic polypropylene. Isotactic polypropylene has various spherulitic forms, based on birefringence measurements and determined by crystallisation conditions [5], and these may be related to the

crystalline structures [6, 7]. But although one can say that the usual spherulitic form is a mixture of Types I and II (using Keith's nomenclature [5]) and that the unit cell is monoclinic from micro-X-ray diffraction patterns, it is not known what form the crystallites take when incorporated in a spherulitic array. Density measurements show that semi-crystalline polymers usually have crystallinities of the order of 70 to 80% and, since small angle X-ray scattering is possible, there must be delineation between the crystalline regions and the amorphous regions within the spherulites. This discrete scattering suggests that one cannot treat spherulites as complete crystals with large numbers of imperfections; in other words, there is a level of order between the individual molecules and the spherulite. The commonest proposal is that there is some kind of radiating fibrillar array, which for polyethylene in particular is refined down to lamellae having a periodic twist [8]. There is another basis of structure which is a semicrystalline, helically-coiled fibril [9] and which explains many of the X-ray and mechanical properties well. However, it suffers from the disadvantage that lamellar structures have been seen on many electron micrographs [8], though again these pictures were obtained from rather specialised conditions, usually from dilute solutions of the polymer under investigation, and it may be that, in the bulk melt, growth does not follow the structural lines that the nuclei would suggest.

The polymer chains have been shown to be approximately tangentially oriented in the spherulite [10]; a fact which fits both structural theories, but which must also be incorporated in a

structural model for polypropylene which has a positive birefringence in the Type 1 form. The birefringence for a long object is defined as the difference between the refractive indices along the long axis and along the short axis: the birefringence being positive when the long axis has a greater refractive index than the short axis and negative when the long axis exhibits the smaller refractive index. In the case of spherulites, the long axis is defined as the radial direction and the short axis as the tangential direction with respect to the spherulite as a whole. The difficulty for Type I polypropylene is that, from optical considerations, a unit cell with the *c*-axis (the chain direction) tangential to the spherulite should result in a negatively birefringent spherulite [11]. The birefringence characteristics of polypropylene can be explained by there being two sets of lamellae exhibiting a cross-hatched pattern [12]. Binsbergen and de Lange [12] have shown that the dimensions of the monoclinic unit cell are such that an epitaxial fit is possible along the (010) plane which would result in a dendritic type of growth with two sets of lamellae crossing each other edge-on at an angle of approximately 80°. This cross-hatched structure has been seen both in quadrites [13] and in etched spherulites [14]. The increase in epitaxial misfit with temperature also accounts for the more negative birefringence at the higher temperatures of crystallisation [12] and so, conversely, by measuring the birefringence it should be possible to estimate the proportion of tangential lamellae. This in turn should have some bearing on the deformation processes, for the structure should become less anisotropic with an increasing proportion of tangential lamellae.

The tangential strength of polyethylene apparently depends on three factors: the interweaving of radial lamellae, the presence of interlamellar tie molecules and the intermolecular van der Waals forces. Polypropylene should possess greater strength in a direction tangential to the spherulites than polyethylene, because of the lamellae actually running in this direction, though at the same time the fact that these lamellae are not radially oriented will weaken the radial direction.

2. Experimental

This paper reports some observations on the deformation of fully-spherulitic bulk polypropylene using microtomed sections cut from the tensile specimens.

The tensile specimens were cut from discs of isotactic polypropylene 5 mm thick and 110 mm in diameter. The discs were compression moulded from Propathene granules supplied by ICI Plastics Division, Welwyn Garden City, Herts, UK, using a specially-designed press [15]. The Propathene granules were heated to 523 K and then cooled at a rate of 0.04° per sec to room temperature, to give a disc with an average spherulite diameter of 0.16 mm. The tensile specimens were cut out using a high speed air-driven router and then polished with fine emery paper.

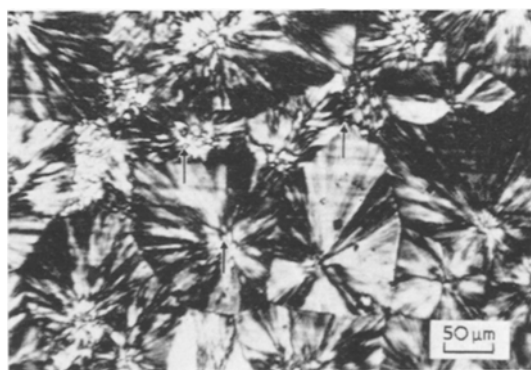


Figure 1 The original spherulitic structure of the specimen prior to tensile tests. Transmission optical micrograph with polarised light.

3. Results

The spherulites in the moulded discs were found to have a positive birefringence of the monoclinic Type I form and, under polarised light, were seen to have a birefringently irregular structure (fig. 1). This is usually the case with polypropylene; the regular "maltese cross" pattern found with polyethylene and other more perfectly crystallising polymers does not appear until after some annealing and recrystallisation process [16]. This may be due to the continuous cooling, as opposed to a single crystallisation temperature, giving rise to changing crystallisation kinetics and growth habits. There is a very definite radiating array visible in spherulites which have been sectioned diametrically. The granular appearance of some spherulites is due to sectioning across a chord and viewing the "fibrils" end on. Around some spherulite boundaries and in the granular regions, there are black lines which do not appear to change in intensity upon specimen rotation. This signifies

that these regions are optically isotropic. This can be explained if the regions are amorphous, or if they are areas of zero birefringence due to the relative proportions and directions of sets of lamellae.

The strain rate for the tensile tests was chosen to be low enough to give cold drawing, whereby the neck initially formed in the gauge length stabilises by strain hardening and propagates to the shoulders before finally fracturing in a fibrillar fashion. The tests were stopped at fixed strains and the specimens removed after reversal of the crosshead to zero stress. The plastic strain was measured and so the elastic strain calculated. It was found (fig. 2) that there was very little

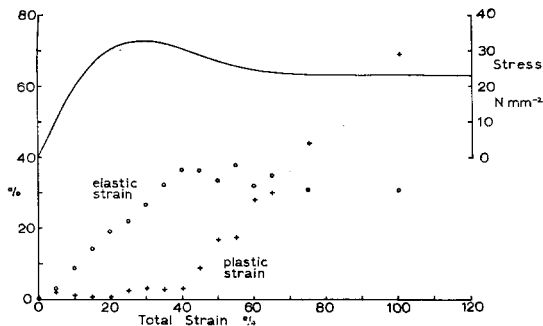


Figure 2 Graph showing typical variation of elastic and plastic strain with total strain of the specimen, with superimposed typical normal stress/strain curve.

plastic deformation up to 25% total strain, which corresponded to the maximum normal stress, the yield stress. After this point a few per cent plastic strain was measurable until the total strain was 40%, when the elastic strain reached a maximum of 35% and the plastic strain started increasing linearly in step with the total strain. There was no visible change in the microstructure up to the yield stress, then some strain whitening became visible on a macro scale and at the same time there was a change in the microstructure, but at this stage very little and only detectable under unpolarised light. It took the form of what appeared to be small cracks running along spherulite boundaries and in a few cases running into spherulites (fig. 3). In nearly all cases these cracks were aligned approximately perpendicular to the applied stress.

At greater strains, when the specimen was starting to neck, these "cracks" took a more random form though still running perpendicular to the applied stress. Under polarised light they were seen as bands with dark lines around the

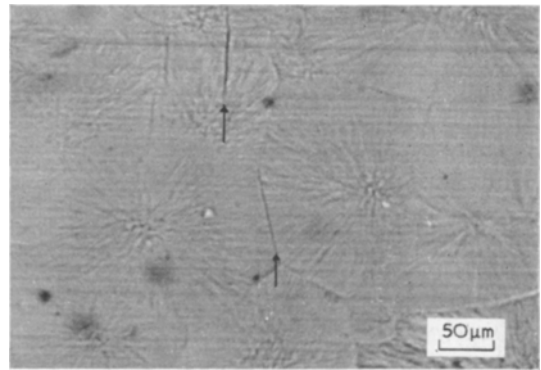


Figure 3 Cracks visible with unpolarised light in the spherulitic structure after 40% total strain. Strain direction horizontal.

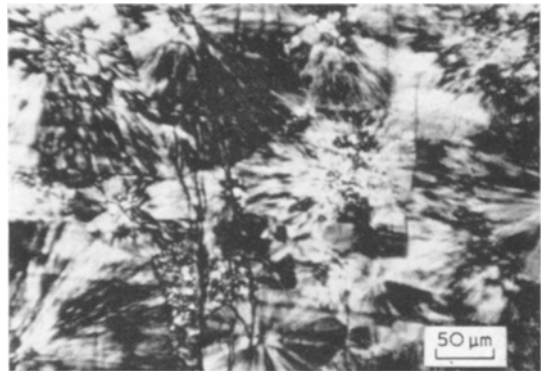


Figure 4 Oriented polymers in random bands running normal to the stress. 50% total strain, 17% plastic strain.

edges (fig. 4), the dark lines being non-birefringent and having a similar appearance to the cracks seen at lower strains. The molecular orientation within these bands, determined from the birefringence, was found to be in line with the applied stress.

As the strain in the necked region increased to approximately 60% overall, striations appeared within the spherulites (fig. 5). These were in radial cones oriented parallel to the applied stress and were seen as black arcs essentially perpendicular to the stress. Again, as the strain increased, these opened out and had a similar appearance to the random deformations previously mentioned: i.e. bands with dark lines around the edges. At the same time the spherulites as a whole became more elongated with an associated decrease in width.

The neck was well formed at 75% total strain and 130% average strain in the actual necked



Figure 5 Striations in necked region in the form of arcs perpendicular to and oriented along the stress direction. Stress direction horizontal at 60% overall strain and 70% maximum strain in necked region.

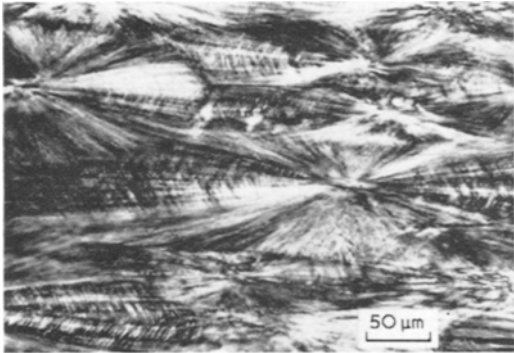


Figure 6 Chevron and granular regions in spherulites in specimens deformed 75% overall with 130% strain in neck region. Stress direction horizontal.

region. The microstructure (fig. 6) showed elongated spherulites with relatively sharp longitudinal boundaries, but more diffuse transverse boundaries perpendicular to the stress. The presumed inhomogeneous deformation regions still lay in narrow cones along the stress direction, but the arcs now had the appearance of chevrons implying a greater deformation along the central axis. At the same time these arcs were continually increasing in number with the strain. The spherulite segments running perpendicular to the applied stress were also becoming more granular in appearance, losing the radiating fibrillar structure of the rest of the spherulite.

At this stage the necked region was completely white and opaque when viewed in bulk. This effect was seen microscopically in the increased opacity of the sections, due to the increased

birefringence and the birefringence alignment along the stress axis. This is an orientation effect, both of lamellae and of microvoids. The spherulites could still be detected as drawing continued in spite of the increasing lack of contrast as the orientation increased. It was found that some drawing in the necked region still continued while the neck propagated along the gauge length until fully fibrillar fracture occurred. Even at 500% total strain the radial structure was visible (fig. 7), mainly in the originally 45° directions. The rest of the visible structure was granular. In the regions where inhomogeneous deformation had been seen, along the applied stress direction where the arcs were first seen, the specimens were less birefringent and so darker, showing that these were relatively undrawn areas.

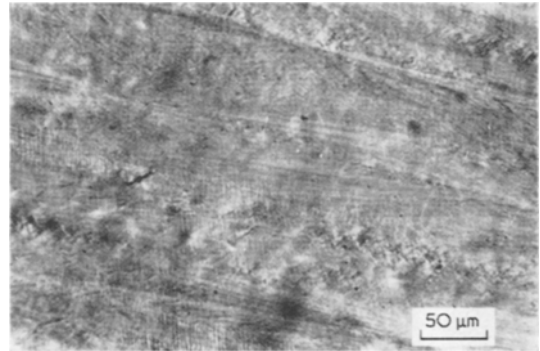


Figure 7 Remnants of radiating spherulitic structure at 500% total strain, 620% maximum strain. Darker, granular areas still contain parts of the original, undeformed, lamellar regions. Stress direction horizontal.

A typical specimen was pulled to fracture and the strain measured during extension using 1 mm spaced ink marks. The maximum strain measured was plotted against the overall (or total) strain of the specimen (fig. 8) and found to be divided into several distinct sections with different gradients. These have been related to suggested stages in the deformation process.

One difficulty encountered as the extension increased, was premature fracture at the shoulder. Presumably the cold drawn volume attempts to propagate up the radii at the ends of the gauge length and creates a sharp angle between the fully drawn and the undrawn material. Study of the fracture surface showed a large cavity in the centre of the specimen (fig. 9) and sectioning specimens before fracture occurred showed voids forming at the ends of the long axes of

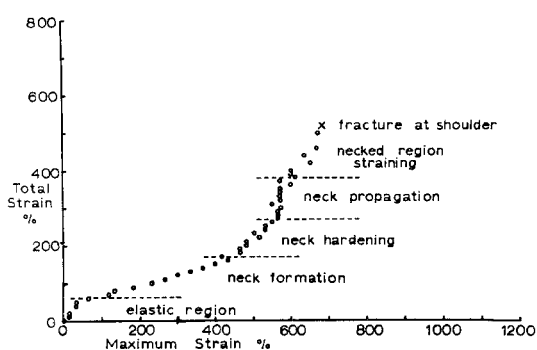


Figure 8 Graph showing different stages in the deformation with relation to the strain. (A different specimen from that described in fig. 2)

deformed spherulites in this region. Increasing the radii at the ends of the gauge length was found to have little beneficial effect.

It should be emphasised that the surface finish of tensile specimens is important: any surface blemish over the gauge length easily propagates and causes premature fracture.

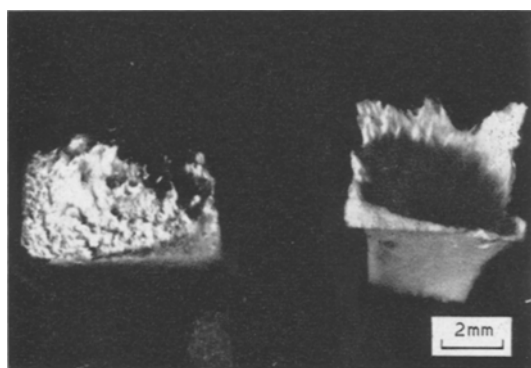


Figure 9 Appearance of fracture surfaces when cavitation causes premature failure at the shoulder.

4. Discussion

A study of the original material under polarised light showed the presence of thin black lines of non-birefringent material, especially noticeable when viewing spherulites sectioned across a minor chord (see fig. 1, some black lines are indicated by arrows). These separated areas of different orientation of the birefringence long axis, the actual values of the birefringence being similar. This suggests that the "fibrils" are grouped together in quite large bundles (10^{-5} m, $10 \mu\text{m}$ diameter), each bundle having a similar crystal orientation within it. The diametrically sectioned

spherulites show that these bundles extend to the centre of the spherulite.

At 2% plastic strain and a stress of 30 N mm^{-2} , the transverse dark lines appeared between spherulites (fig. 3). These lines were probably a series of voids along the spherulite boundaries; a conclusion reached from previous work carried out in these laboratories [17] using a scanning electron microscope. The apparent lack of birefringence here would be due to the small amount of material actually passing from one spherulite to another and to the amorphous material rejected during spherulite growth.

The first signs of the arcs along the tensile direction appeared at approximately 17% plastic strain and 37 N mm^{-2} stress (fig. 4) and at the same time more random inhomogeneous deformations occurred perpendicular to the stress, having the form of black lines around oriented material. The black lines around these areas signify optically isotropic regions. This is probably an amorphous "unravelling" portion where the original, spherulitic, crystalline structure is being reformed to a fully oriented structure with the molecular chains lying along the stress direction. Any explanation for the formation of the arcs and for the subsequent characteristics of deformation under further straining must depend on the particular interpretation of the internal structure. We now present our interpretation.

It would appear that the type of deformation is dependent on the angle that the fibrils make to the direction of stress. This in turn means that the structure is anisotropic. It was noted that there were three separate regions: the regions where arcs formed, the regions where fibrils persisted throughout the deformation process, and the regions which had granular appearances at later stages in the deformation (fig. 10). The arc forming region covered an angle of about 25° , while the region which later became granular, which lay perpendicular to the stress, covered an angle of about 120° when it first appeared.

Using the relation [12] for the birefringence

$$\Delta n = (n_c - n_a)(t - r/2)$$

where r and t are the fractions of radial and tangential lamellae respectively and the birefringence of the unit cell ($n_c - n_a$) is $+0.067$ [11], the ratio of radial to tangential lamellae from the measured birefringence of $+0.007$ is 6:4. The evidence for polyethylene shows a fully radial orientation of lamellae within a spherulite [3]

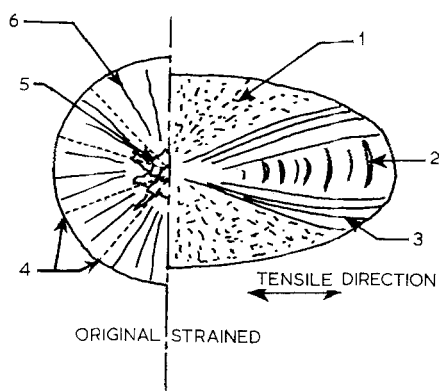


Figure 10 A line diagram showing the different deformation regions.

Key: 1. Granular region. 2. Arcs, or chevrons at higher elongations. 3. Unaltered radial fibrils. 4. Positions of deformation boundaries. 5. Black-ringed region. 6. Radiating fibrils.

and, on deformation, drawing occurs along diameters perpendicular to the stress, suggesting that the weakest regions are between the lamellae. Returning to polypropylene: if one assumes the maximum stress to be located down the long axis of the deformed spherulite, then yield will first occur along the faces of tangential lamellae lying across this axis. This yielded region will act as a craze and propagate approximately perpendicular to the stress. In fact it will follow the lamellae faces and hence the curvature noted in the arcs seen, since the spherulite grows from the melt as a sphere and the tangential lamellae will grow on the face of this sphere. One would not expect the tangential lamellae to be of any great length, otherwise the overall curvature of these arcs would not be maintained. Since the growth direction of spherulites is predominantly radial, the tangential growth will be limited by impingement on neighbouring radial elements, hence, though there may be a large proportion of tangential lamellae growing in a dendritic fashion, they will be short, unlike the radial lamellae which can grow outwards unhindered. This would also explain why a radial pattern is seen in the spherulites, rather than a series of concentric shells. After yielding has occurred, the number of arcs does not increase considerably, since subsequent pulling-out and reorientation of the molecular chains within an arc is easier than creating a new arc in another region. The radial lamellae will tend to hold together the undeformed regions. Extension of the spherulite by several hundred per cent increases the number

of arcs. The originally-deformed regions strain-harden, because of the difficulty of pulling out further chains entangled in the amorphous boundary of the oriented region, and consequently it is easier to form new arcs. However, even at 500% extension, the birefringence of the structure between the arcs in these cones of the spherulites is still positive and of a value the same as that of the original structure. This shows that, although the rest of the specimen is negatively birefringent as would be expected (considering the long axis to lie along the stress direction), the regions between the oriented arcs still maintain the crystal structure of the original spherulite.

As mentioned previously, the fibrils lying at an intermediate angle to the stress maintain their form throughout the deformation process. Thus there can be no large-scale inhomogeneous disruptive force. A deformation process which would account for this lack of disruption is rotation of lamellae about axes normal to the stress, so as to lie more in the direction of stress, accompanied by inter-lamellar sliding. The sliding being a shear process along the weak interlamellar boundaries. Measurement of the birefringence of moderately drawn specimens gives a value of -0.03 relative to a long axis along the stress direction and this, using the aforementioned relationship, gives a fraction of lamellae along the draw direction of just under unity. In other words, in this region the structure consists almost entirely of lamellae stacked along the draw direction. There is an angle between 10 and 15° from the stress direction at which there is a changeover from the arc type of deformation to the shear deformation. Once this boundary has formed it remains static relative to the spherulite boundary, elongation of the spherulite simply decreasing the angle the deformation boundary makes with the stress direction. The radiating fibrils were straight initially, but it was found that, after a few hundred per cent strain, as well as being rotated they also became slightly curved, showing that deformation was not completely affine. However, they did not have the curvature that might be expected from the type of deformation suggested by Wilchinsky [18], which assumed concentric shells deforming with constant separation, though the centres of spherulites were seen to be highly elongated as this model predicts. It is very probable that the region close to the spherulite nucleus has a different structure from the more

regular outer regions and so, at the centre, one may expect a different yield pattern, hence the greater elongation than an affine process would give.

The region of the spherulite perpendicular to the stress maintains a fibrillar appearance up to quite high strains (about 150%), but at about 100% strain (70% total strain for the specimen) the fibrils become indistinct and as the strain increases a granular appearance is attained over an included angle of about 120°. This granulation occurs at a higher stress than that required for arc formation; in fact about twice as high. This means either that the tangential direction is much stronger than the radial direction, or that there are large variations in the stress field within a spherulite.

An explanation based on the former proposition is that instead of yield occurring along one surface, as in the arcs, it is occurring generally on a very fine scale so that the initial stages are not detected. We believe that interlamellae strength is low, so this is the most likely point of yielding, and since in this region the majority of lamellae run perpendicular to the applied stress, there will be a higher proportion of weak regions lying across the stress compared with the other parts of the spherulite. By similar reasoning, the number of yielded regions formed will not be as limited in plane, or number, by the lamellae running parallel to the stress. Hence there will be an overall, ostensibly homogeneous, break-up of structure. However, if deformation were based on this criterion alone, it would be very sensitive to changes in the proportions of the differently-oriented lamellae, and one would expect an occasional granular break-up where one normally has the arc type deformation, and vice versa, since crystallisation processes would not be identical throughout a specimen, resulting in varying fractions of radial and tangential lamellae. Even in other specimens subjected to different cooling conditions, there was no great departure from the visible deformation processes being described and so it is probable that some other condition is necessary to produce the observed effects.

The difference in deformation processes is then likely to be due to a variation of stress within the spherulite, with the highest stress being concentrated along a diameter running parallel to the applied stress. It is likely that the molecular chains are aligned across the spherulite boundaries, so very little strain will produce a relatively rigid bond between the spherulites in the direction

of stress. If one assumes the presence of fairly-continuous radial lamellae, then these will provide rigidity down the stress axis, while, at an angle to this axis, fairly easy interlamellar slip will reduce the stress acting on the regions lying perpendicular to the stress, hence there is an inhomogeneity in stress concentrations.

The apparent lack of any appreciable deformation along longitudinal interspherulitic boundaries suggests that neighbouring spherulites have little effect on the deformation. In other words, there is little interspherulitic movement. However, there is one important exception to this generalisation, and that is when the drawn/undrawn boundary reaches the end of the specimen gauge length and propagation of the drawn section stops due to the increasing specimen cross-section. There is a very sharp radius produced, especially when the cross-section of the drawn region is reduced by further straining, and at the same time there is a rapid change from a region of undrawn spherulites to the drawn spherulites. Associated with the elongation of spherulites there is a reduction in cross-section perpendicular to the stress, an effect of small consequence when the specimen is unconstrained laterally. However, in the neck region the undrawn material acts as a constraint to those spherulites trying to contract laterally, immediately next to it. Also, the spherulites at the specimen surface will have been strained more and are consequently more rigid than those at the centre and so less likely to yield inwards to allow for the contraction. The nett effect is that a hydrostatic tensile stress is built up in the centre of the specimen in the neck region and this results in voids forming at the weakest points. These are the interspherulite boundaries, usually where three spherulites meet, for previous experiments [17] have shown that this is where the greatest concentration of microvoids is formed on cooling. The reason why these voids are not formed under the normal drawing is then due to the triaxial stress system existing in the neck as opposed to a uniaxial system existing in the cold drawn region. This void formation, which eventually leads to fracture at the neck (fig. 9) has proved difficult to avoid: increasing the radius at the ends of the gauge length only causes the neck to move farther past the gauge length before finally stopping and again voiding.

5. Conclusions

As far as is known, isotactic polypropylene is

unique in having a unit cell whose similar a and c repeat distances permit the epitaxial cell fit which gives rise to a cross-hatched structure. The fact that isotactic polypropylene has such a structure has been known for some time [19], but its importance has not been fully realised. We believe that the cross-hatched structure can be used to explain some of the characteristic properties of polypropylene, for example the anomalous birefringence when compared with other spherulitic polymers [12]. In this paper we show that the deformation of polypropylene spherulites may be explained in terms of the two sets of lamellae and, also, by postulating that planes of weakness lie along lamellar faces and that the spherulite itself is an inhomogeneous stress system.

The greater degree of isotropy implied by the cross-hatched structure would also explain the good creep and fatigue properties of polypropylene. In both cases the tangentially oriented lamellae will bind the radial lamellae together and impede crack growth between the lamellae. As was noted in studying the original undeformed spherulites, a minor chord across a spherulite showed the fibrils to be grouped in discrete bundles with similar orientations (as deduced from the birefringence) with thin boundary layers between the bundles. These black boundary layers are probably due to a difference in orientation of lamellae between adjacent bundles, as in crystal orientation between grains in a metal. The amorphous layers would then act as easier paths for cracks to follow and, since they are oriented radially, one would still anticipate trans-spherulitic cracks. However the branching of lamellae during growth will give strength to these regions. Lamellar branches will cross into adjacent bundles of lamellae and become physically intertwined.

Polypropylene also has good environmental stress cracking resistance. The explanation for this type of cracking is that the environment plasticises the material at the crack tip allowing easy removal of chains and so crack propagation. This would be expected to occur most readily between lamellae where there are only tie molecules, the lamellae being inherently more tightly bound and so stronger. The existence of lamellae running across the gaps between radial lamellae would then be a strengthening factor.

It has been found [17] that, in general, the larger the spherulite size the fewer are the

interspherulitic links and this results in a more brittle, low strength material because of the easy crack propagation along spherulite boundaries. Consequently, if one can produce small spherulites, the internal spherulitic structure is of much greater importance. However, the faster cooling rate required for the greater nucleation rate to give smaller spherulites also results in a coarse, poorly formed spherulitic structure which does not give the best mechanical properties. For optimum mechanical performance, the overall requirements would then seem to be that one requires small spherulites, to reduce the incidence of spherulite boundary weakness, combined with a refined, well formed, cross-hatched, lamellar structure.

Acknowledgement

We thank the Science Research Council for the award of a Studentship to J. L. Way.

References

1. L. BARISH, *J. Appl. Polymer Sci.* **6** (1962) 617.
2. R. J. SAMUELS, *J. Polymer Sci.* **C20** (1967) 253.
3. I. L. HAY, and A. KELLER, *Kolloid-Z* **204** (1965) 43.
4. T. ODA, S. NOMURA, and H. KAWAI, *J. Polymer Sci.* **A3** (1965) 1993.
5. H. D. KEITH, F. J. PADDEN JR., N. M. WALTER, and H. W. WYCKOFF, *J. Appl. Phys.* **30** (1959) 1485.
6. G. NATTA and P. CORRADINI, *Nuovo cim.* **15** (1960) 40.
7. A. TURNER-JONES, J. M. AIZLEWOOD, and D. R. BECKETT, *Makromol. Chem.* **75** (1964) 134.
8. P. H. GEIL, "Polymer Single Crystals" (Interscience: New York, 1963).
9. L. B. MORGAN and P. G. STERN, *Polymer London* **9** (1968) 375.
10. A. KELLER, *J. Polymer Sci.* **17** (1955) 351.
11. D. A. KEEDY, J. POWERS, and R. S. STEIN, *J. Appl. Phys.* **31** (1960) 1911.
12. F. L. BINSBERGEN and B. G. M. DE LANGE, *Polymer London* **9** (1968) 23.
13. J. A. SAUER, D. R. MORROW, and G. C. RICHARDSON, *J. Appl. Phys.* **36** (1965) 3017.
14. G. W. BAILEY, *J. Polymer Sci.* **62** (1962) 241.
15. J. M. GREIG and J. R. ATKINSON, to be published.
16. R. G. CRYSTAL and D. HANSEN, *J. Appl. Phys.* **38** (1967) 3103.
17. J. L. WAY and J. R. ATKINSON, to be published.
18. Z. W. WILCHINSKY, *Polymer London* **5** (1964) 271.
19. F. KHOURY, *J. Res. Nat. Bur. Stand.* **70A** (1966) 29.

Received 14 September and accepted 27 October 1970.