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On cellulation and banding during crystallization of a linear-low-density polyethylene from linear nuclei

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Abstract

Linear-low-density polyethylene shows novel features, in addition to those found for the linear polymer, when crystallized as row structures from linear nuclei. There is the same division between disordered and ordered (inclined) fold surfaces for faster and slower crystallization, respectively, with the changeover occurring at a lower temperature and for a longer time than the linear polymer. The first new feature is that the kinetics of slower crystallization, with inclined fold surfaces, are non-linear, slowing as segregation proceeds, then recovering approximately to the initial value as cellulation sets in. Two patterns of cellulation, with associated kinks in the kinetic curves, have been observed. At all temperatures, a morphology similar to that encountered in linear polyethylene occurs when lamellae have become normal to the nucleating fibre. Beyond this, at 123 °C, is a coarser cellulation related to the onset of banding. This banding, the second new feature, contrasts with that in linear polyethylene which only occurs for faster crystallization, when the driving fold-surface stress is caused by the initial disordered packing itself. For slower crystallization in the linear-low-density material, fold-surface packing was initially ordered but surface stress is created, which banding helps relieve, when isothermal lamellar thickening is restricted by branches brought into the fold surfaces. © 2002 Published by Elsevier Science Ltd.

Keywords: Polyethylene; Cellulation; Banded growth

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1. Introduction

The study of polyethylene grown on high-melting fibres as linear nuclei, to form row structures in the absence of stress, has brought new insights into polymeric crystallization from the melt. In the linear polymer, crystallization divides into two temperature regions according to whether fold surfaces are or are not ordered before the next molecular layer is added to a growing lamella [1]. In the lower range, for *faster crystallization*, ≤126 °C in the previous experiments, lamellar surfaces are initially *perpendicular*, i.e. {001} and norma100 l to the nucleating fibre, but the dominants among them become non-planar with increasing radial distance and isothermal thickening, developing S-profiles when viewed down **b**, the growth direction [2]. This is the mode associated with banded growth. On the other hand, *slower crystallization* in the higher temperature range, ≥127 °C, has lamellae with *inclined*, approximately {201}, fold surfaces oriented from the start at ± ~ 35° to the fibre axis. In this case, banded growth does not occur but there is a novel kinetic effect when this initial state gives way to a faster-growing one in which fold surfaces are still ~{201} but now with lamellae well-separated and normal to the fibre. The change was interpreted as due to the initial occurrence then loss of internuclear interference when one molecule is able to crystallize, in Regime I, simultaneously on two lamellae.

Linear-low-density polyethylenes with as many as 21 butyl groups per 1000 C atoms, were previously found to show the same division from initially perpendicular to initially inclined ~{201} fold surfaces [3]. This occurs at a lower temperature than, but at a comparable growth rate to, the linear polymer. In this paper the study of a further linear-low-density polyethylene, Sclair 14B (duPont, Canada), a film grade of density 0.932 g cm^{-3}, with 1.9 wt% of butene-1 as comonomer, i.e. 4.8 ethyl branches per 1000 C atoms, has led to two new findings in the slow growth mode. First, although the morphology of ordered surfaces initially follows the same sequence as for the linear polymer, isothermal radial growth rates are now not linear. This is

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because of segregation of more branched molecules and/or sequences and ensuing cellulation. Second, in a new effect, banding occurs late notwithstanding the initially ordered fold surfaces. Previously banding has only been observed for faster growth and disordered fold surfaces.

In this system growth rates generally decline with greater radial distance as segregation increases but show systematic increases revealed as upward kinks in plots of growth length against time, with growth rates reverting approximately to or, at the highest temperature, probably exceeding the initial value. In both cases increased growth rates correlate with the appearance of a cellulated morphology implying reduced segregant concentrations at the growth interface; the loss of internuclear interference may also be a factor. The first kink occurs when inclined lamellae have become normal to the nucleating fibre with a morphology resembling that seen for segregation of shorter molecules in the linear polymer. A coarser morphology is associated with the second kink, found when banding begins to develop, with dominant lamellae adopting shallow S-profiles behind the growth front where fold surfaces remain inclined and planar. This late banding for slow crystallization is suggested to relieve surface stresses created when isothermal thickening is restricted by branches brought into the fold surfaces: for faster crystallization the stress is caused by the initial fold-surface packing itself.

2. Experimental

Specimens containing high-melting polyethylene fibres as linear nuclei were prepared using flat discs of the polymer, ~20 × 30 mm across and either ~1 mm or 60–80 μm thick, produced from the appropriate quantities of pellets by melt pressing, between a clean 13 μm thick Kapton sheet and a microscope slide, on a Kofler hot-bench at 150 °C for 5 min. At the end of this time each disc with the Kapton sheet and the microscope slide was quenched by placing on a large metal plate. After removal from the microscope slide it was cut into small squares of ~5 mm side which were stored in labelled jars ready to prepare a fibre/polymer composite.

A typical specimen containing about seven highly oriented Tekmilon™ polyethylene fibres (Mitsui) within a matrix of the chosen polyethylene was prepared from fibres sandwiched between two discs, one 1 mm thick, the other 60–80 μm thick, both prepared as above. The two discs were melted together for 3 min on the Kofler hot bench at 133 °C, then moved to a Mettler FP82HT hot stage for a further 5 min at 133 °C before cooling to the required crystallization temperature using a Mettler FP90 central processor. The composite specimen was quenched in ice–water mixture after the selected time of crystallization.

All specimens were etched in a 2% (w/v) potassium permanganate solution in 10:4:1 parts by volume of concentrated sulphuric acid, 85% orthophosphoric acid and distilled water, respectively, for periods up to 4 h. Shorter times, monitored by Nomarski reflection optics, exposed surfaces as in Fig. 1, through, and parallel to, the row structures but outside (in the limit tangential to) the nucleating fibres. Such surfaces give what we term *radial views*: down the growth axis, **b**, (which is radial to the row) along a central line and out to the edge of the row at either side. The precise information varies according to the particular chord of the near-circular cross-section of the row exposed. This ranges from the narrowest chord, which shows essentially just the view down the growth axis, through to the start of growth being visible at the interface with the fibre in sections tangential to it. In those cases when the fibre is slightly inclined to the etched surface, views at different distances from the fibre may conveniently be observed simply by moving along the length of the exposed row. Longer etching times were required to expose *diametral sections*, deeper than those of Fig. 1 and passing through the centre of the nucleating fibre or nearly so; these were used to measure growth length, with some twenty measurements taken per point. Etched specimens were either coated with gold prior to examination in the scanning electron microscope, SEM, or replicated by a standard two-stage process for transmission electron microscopy, TEM. All the micrographs in the figures are of etched surfaces recorded in the SEM.

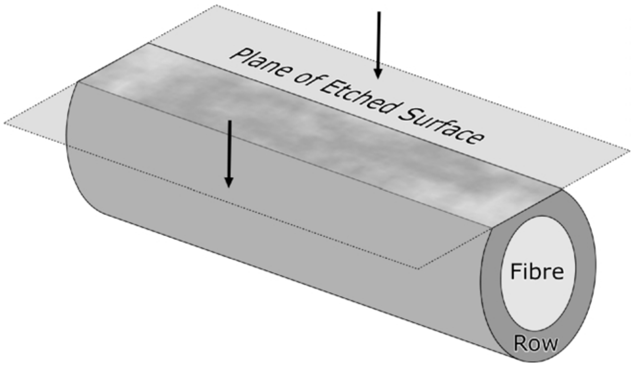

Fig. 1. Schematic of specimen geometry after material has been removed, by etching, in the direction of the arrows.

3. Results

The morphology of row structures of Sclair 14B linear-low-density polyethylene shows a clear change between 119 and 120 °C. For faster growth at 118 °C and 119 °C (temperatures at which the polarizing microscope shows banding with an interval of 2–3 μm from the fibre to the first dark band) the SEM shows that lamellae form normal to the nucleating fibre (Fig. 2(a)) whereas for slower growth at 120 °C (Fig. 2(b)) they and their fold surfaces are doubly oriented from the start. This corresponds to the same division observed for linear polyethylene between 126 and 127 °C at a somewhat faster growth rate [1]. However, the

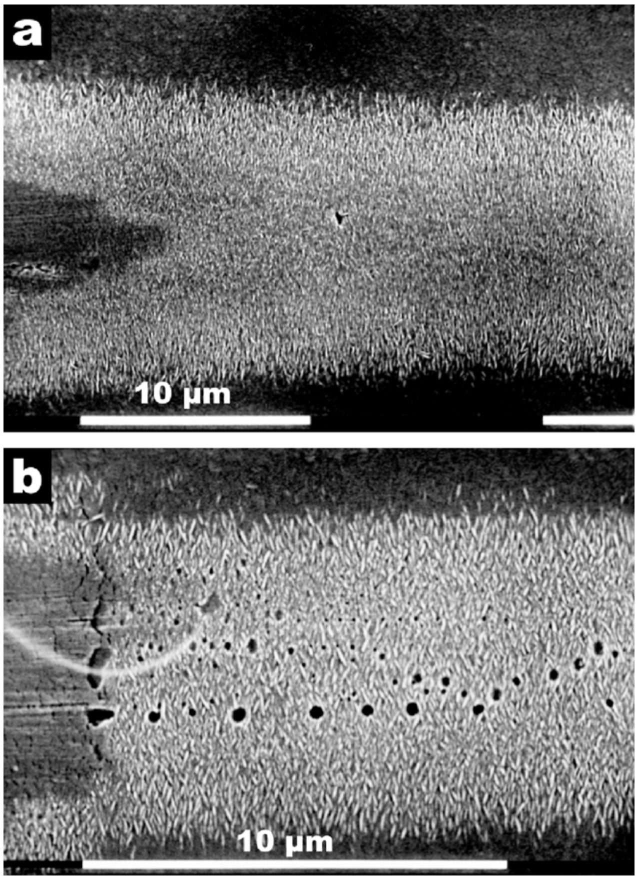

Fig. 2. Radial views of row structures showing the contact zone with the nucleating fibre. In (a) for 30 s at 118 °C, lamellae form perpendicular to the fibre axis whereas in (b) for 2 min at 120 °C, they form with doubly oriented inclination.

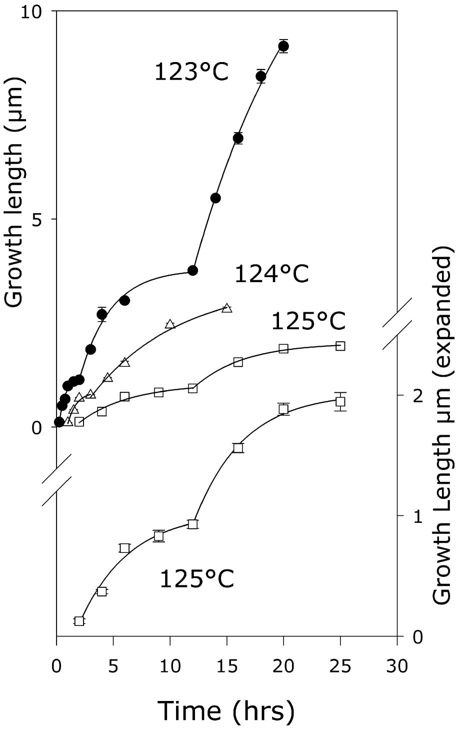

Fig. 3. Plots of growth length against time for the three temperatures indicated. The plot of data at 125 °C is expanded below.

kinetics of slower growth differs from the behaviour of linear polyethylenes. Fig. 3 shows, above, data points for average radial growth length outside the nucleating fibre as a function of crystallization time for 123, 124 and 125 °C together with connecting lines. All three plots show kinks in which growth rates, i.e. the gradients of the plots, slow then recover to similar or, at 125 °C (amplified below), probably greater, values than those at the origins. As will be demonstrated, the first kink at 123 °C and those at 124 and 125 °C, occur when the plane of inclined lamellae has become normal to the fibre and coincide, more or less, with the onset of a cellulation morphologically similar to that seen in linear polyethylene when shorter molecules segregate at the highest temperatures [1]. The second kink at 123 °C correlates with a coarser cellulation linked to banded growth. At 124 and 125 °C it was not possible to observe complete banding free from spherulitic crystallization of the adjacent matrix making the region of a presumed second kink experimentally inaccessible.

Fig. 4 shows both radial and diametral views of rows crystallized at 123 °C for pertinent times in relation to the kinks of Fig. 3. After 1 h. Fig. 4(a) shows, on the right, that inclined lamellae grow from the fibre and, on the left, that an open diamond-shaped texture associated with cellulation is present near the growth front. However, Fig. 4(b), of the corresponding diametral view, shows that leading lamellae are only ~0.1 µm ahead of their neighbours, a distance comparable to their lateral separation so that cellulation is shallow. After 1.5 h crystallization, Fig. 4(c), lamellae at the side of the row, i.e. at the growth front, are now normal to the fibre as are most of those on the left of the micrograph. The diametral view after this time is little different to Fig. 4(b) in appearance or radial extent, attesting to a reduced growth rate. However, after 3 h (Fig. 4(d)) when growth has accelerated, parallel lamellar orientation prevails together with deep cellulation at the growth front in which there are groups of lamellae ~1 µm ahead of their neighbours. Such morphology closely resembles that observed in the linear polymer at high temperatures, e.g. 129 °C and attributed to the segregation of shorter molecules, cf. Fig. 5b of Ref. [1].

The morphology after 4 h shows considerable change in what is the beginning of twisted growth leading to banding. The radial morphology (Fig. 5(a)) is deeply cellulated at the edges while the diametral view (Fig. 5(b)) shows that lamellae have begun to twist away from the perpendicular orientation in both senses, i.e. to left and right in this view, often adopting curved profiles. There is a hint in Fig. 5(a) of a repeating texture along the row and by 12 h when growth has again slowed substantially, dark vertical regions on Fig. 5(c) are present at the narrowest points, i.e. outermost edges, of the row. These separate groups of parallel lamellae

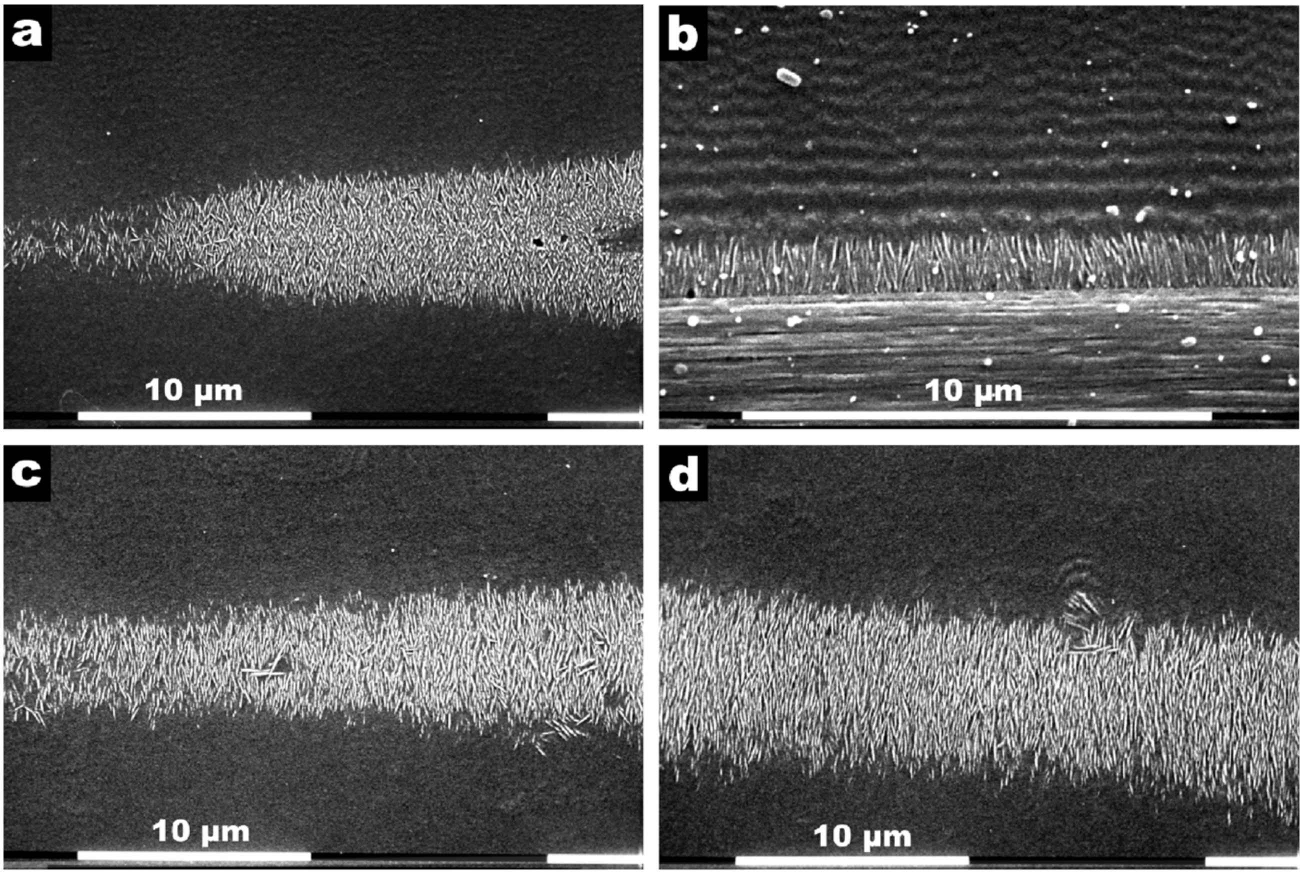

Fig. 4. Row structure morphology at 123 °C for times to the first kink in Fig. 3. (a) Radial view after 1 h; (b) diametral view after 1 h; (c) radial view after 1.5 h; (d) radial view after 3 h.

having planes inclined at ± ~ 45°, either to left or right of the vertical, with a few isolated groups present which are more or less square in cross-section and ~1 μm across. Once growth has accelerated again, after 14 h (Fig. 3) such individual groups of parallel lamellae, now ~2 μm across, are very evident in Fig. 5(d) in what is a pronounced coarsening of the texture.

If one looks deeper into the row by examining a wider chord, Fig. 6(a) shows a texture after 14 h, ~2 μm across, of lamellar groups inclined to left and right of the vertical. In Fig. 6(b), after 18 h, the scale of texture has increased to ~3 μm with curved and S-shaped lamellar profiles prominent. Even when, as in Fig. 6(c) for 14 h, lamellae at the outer edge of the row are all more or less parallel to the row forming the first band, the widths of individual lamellae do not exceed 2–3 μm. The texture of the rows during banded growth may, therefore, be described as one of lamellar bundles which twist, in both senses, around the radius of the row. Such bundles are also apparent from the side in the near-diametral view of Fig. 6(d), also for 6 h.

The correlation of kinks in Fig. 3 with cellulation when lamellae with ~{201} surfaces have become normal to the nucleating fibre is also shown at 124 and 125 °C. After 4.5 h at 124 °C, when Fig. 3 shows that the growth rate is about to accelerate, the right hand of Fig. 7(a) shows that lamellae at the outside of the row are normal to the fibre while deeper into the row, on the left, they are inclined. The overall morphology with micron-sized corrugations along the growth front resembles that of Fig. 4(c) but somewhat developed in the direction of Fig. 4(d). After 15 h, Fig. 7(b) shows the beginnings of twisting growth, akin to Fig. 5(c), but has not yet developed a morphology as in Fig. 5(d) giving accelerated growth.

At 125 °C, Fig. 8(a) shows that, after 12 h, lamellae are only just becoming perpendicular to the row at its outside, beyond a central region of doubly inclined lamellae. This is closer in appearance to Fig. 4(c) than Fig. 4(d), suggesting that more time is needed to produce sufficient cellulation for accelerated growth. In this event, the location of the kink drawn in Fig. 3 would move to the right, thereby increasing the growth rate above that at the start. An increased growth rate would parallel that found for slow growth in the linear polymer, more evident at the highest temperatures, and attributed to the loss of internuclear interference. A possible second kink lies well beyond the experimentally accessible region as, even after 25 h when all dominant lamellae are normal to the row (Fig. 8(b)), there is little intimation that twisting growth has started.

Partial information on chain inclination within lamellae has been obtained from TEM examination of surface replicas and specifically those showing the fracture-induced surface striation along **c**. Detailed study of the range of

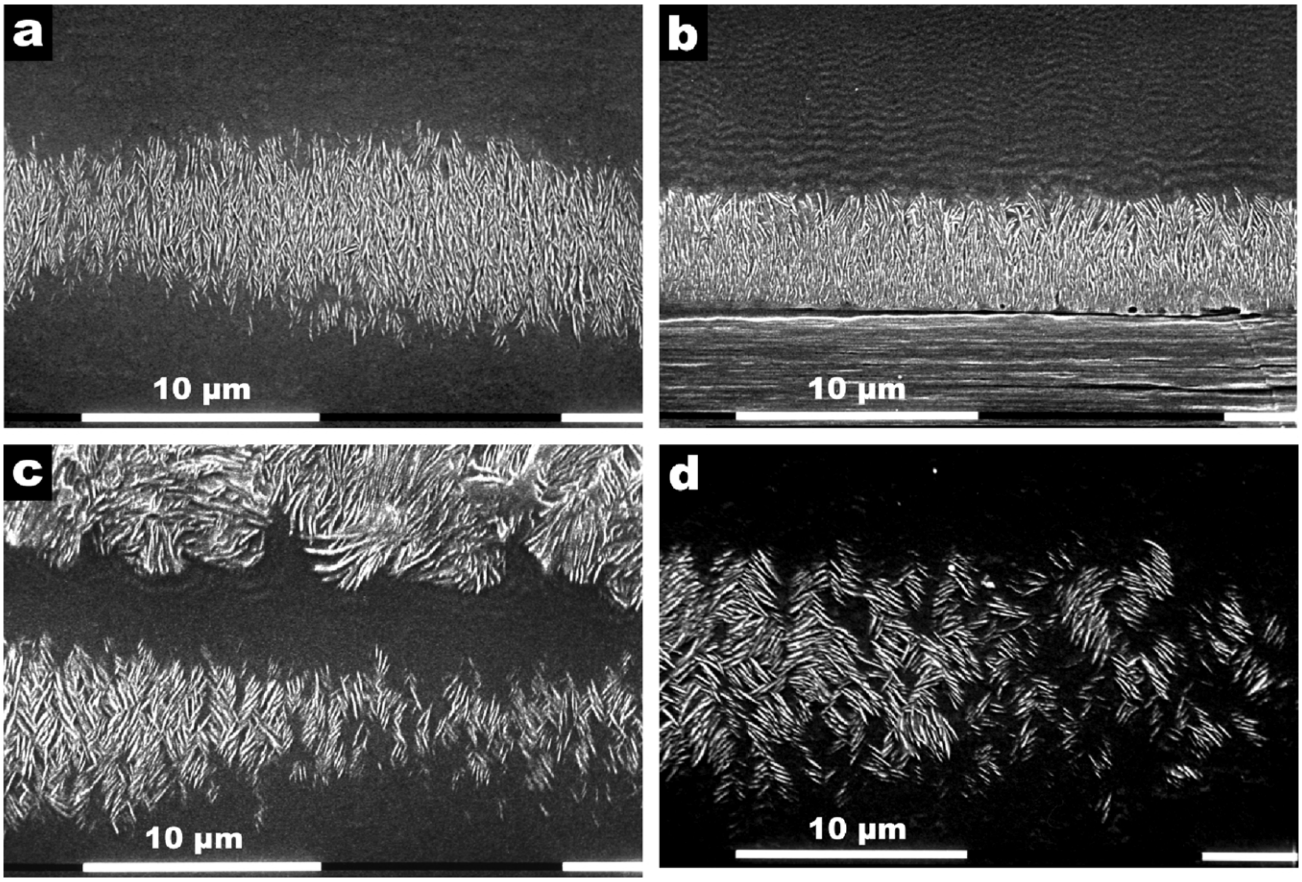

Fig. 5. Row structure morphology at 123 °C for times between the kinks in Fig. 3. (a) Radial view after 4 h; (b) diametral view after 4 h; (c) radial view after 12 h; (d) radial view after 14 h.

morphologies within specimens crystallized for 16 h at 123 °C, has confirmed that fold surfaces remain inclined when lamellae lie perpendicular to the fibre axis as for the linear polymer. This is still the case for planar lamellae at the outside of the row, as in Fig. 6(c); deeper into the row, S-profiles develop. Most of these have the chain inclination decreasing at greater width but in others, to be denoted ‘inverse Ss’, it increases.

4. Discussion

The results of this and previous papers [1–3] are primarily a commentary on the nature of as-formed fold surfaces of polyethylene lamellae and their reorganization during growth. It is well known that polyethylene lamellae tend to adopt inclined fold surfaces, {312} and {314} from solution [4] and {201} from the melt [5], thereby increasing the surface area occupied per fold and reducing surface stress. However, the associated reduction in free energy must be small in comparison to that of crystallizing lamellar interiors so that achieving inclined fold packing cannot be expected always to result from crystallization but rather to occur only when circumstances permit. The most relevant circumstance will be time, as a molecule adds to a lamella, to explore adjacent conformations to those in which it became attached. If this is sufficient, inclined packing will result. If not, a lamella may still reduce its free energy subsequently by improving fold packing as well as increasing its thickness. Isothermal lamellar thickening has long been recorded [6,7], although its details remain rather obscure, but the possibility of fold-surface reorganization has received much less attention. Experiments monitoring the inclination of molecular chains to the fold-surface during growth, as in this paper, reveal such changes directly.

The basic division, as seen previously in linear and linear-low-density polyethylenes [1,3], accords with the above expectation. At lower temperatures with faster growth, lamellae form normal to the axis of the nucleating fibre, i.e. with {001} fold surfaces. They then reorganize during growth, with isothermal thickening, giving S-profiled dominant lamellae and twisting growth producing banding. At higher temperatures with slower growth, ~{201} fold surfaces are present from the start. The change from slower to faster growth was first reported to occur at a rate $>\sim 3\ \mu\text{m h}^{-1}$ in the linear polymer, corresponding to a time to add a single monomolecular layer, 0.5 nm thick, to a lamella of $<\sim 0.5$ s. More detailed investigation, to be published separately, has confirmed that there is a well-defined

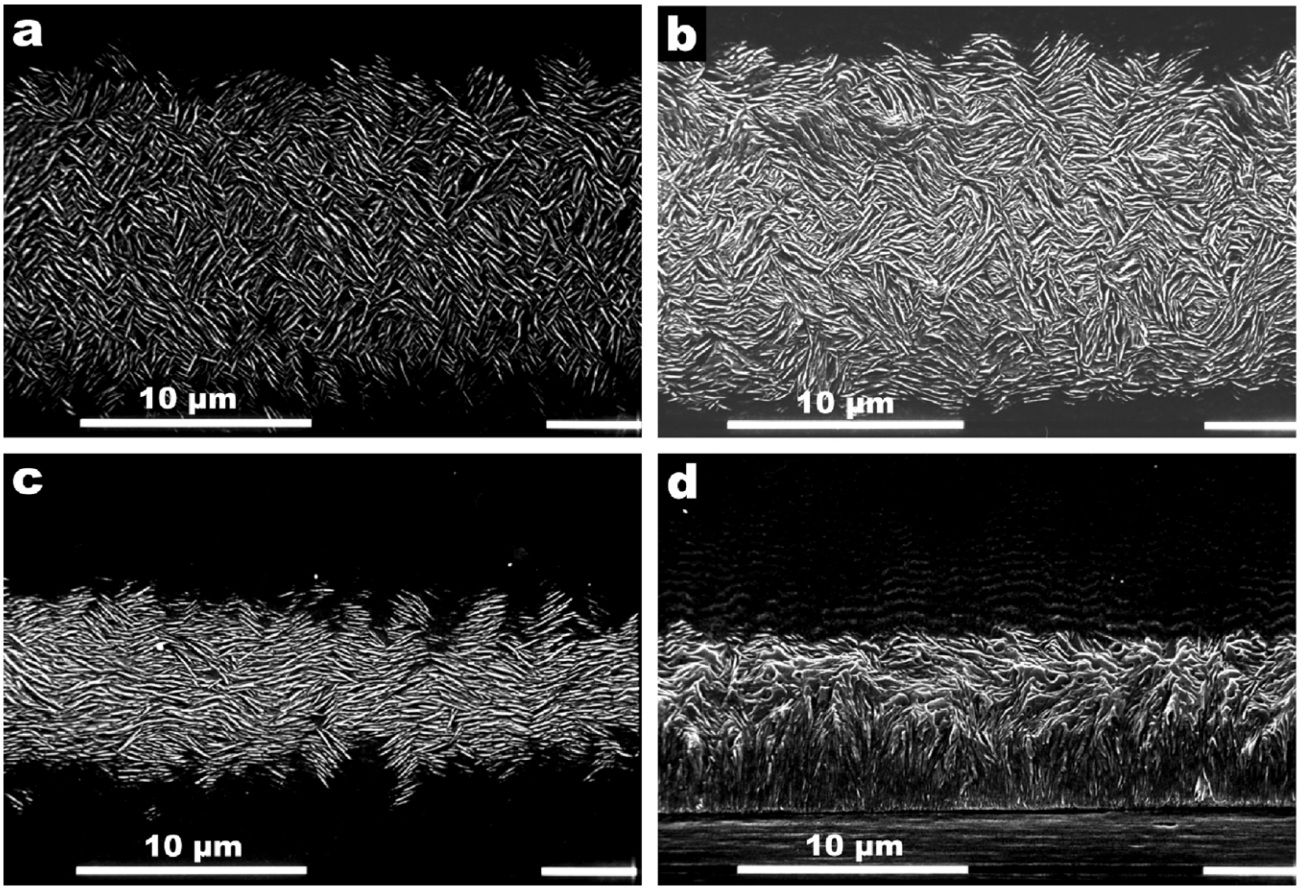

Fig. 6. Row structure morphology at 123 °C for deeper chords and banded growth. (a) Radial view after 14 h; (b) radial view after 18 h; (c) radial view after 16 h near the row edge; (d) near-diametral view after 16 h to show the cellulated relief.

time associated with the onset of fold-ordering for a given polyethylene which increases for longer and more-branched molecules.

The additional behaviour of linear-low-density polyethylene reported in this paper over and above that of the linear polymer must arise from the exclusion of branches from lamellae, the only new factor present. Molecular sequences containing branches closer than the lamellar thickness will be rejected from a growing lamella, slowing the kinetics for statistical reasons and causing segregants to accumulate at the growth interface. Their concentration will increase as growth proceeds, depressing the growth rate further because of the reduced local equilibrium melting point and isothermal supercooling. The return to approximately the original growth rate with deep cellulation (at the first kinks in Fig. 3) indicates that lamellae are again encountering conditions similar to those at the start of growth, having effectively passed through the accumulated segregants which will be concentrated in cell boundaries. The gradual slowing thereafter indicates that this is not a steady state condition but that the segregant concentration at the interface increases again. At this stage the morphologies of the linear and the linear-low-density polymers are very similar even though the nature of the segregants differ: compare Fig. 4(d) with Fig. 5(b) of Ref. [1]. As growth rate does not then fall for the linear polymer, for which transport of molecules to the interface will be similarly changed, it follows that the reduction now observed is a consequence of the reduced isothermal supercooling when branched as opposed to linear molecules are segregated.

The second kink present in Fig. 3 for growth at 123 °C occurs for the same reason as the first but in the changed morphology of banding. Whereas in linear polyethylene banding does not occur when the initial lamellar fold surfaces are ordered, indicating that they are not significantly stressed and do not become so during growth, the observed late banding shows that the situation has altered for the linear-low-density polymer. For this the isothermal lamellar thickening which accompanies growth (as revealed, e.g. by comparing thicknesses in Figs. 4 and 5) will be restricted by excluded branches and introduced surface stress. Measurements of lamellar thickness at the outside of the row, to be published separately, show a linear increase with the logarithm of time. At 123 °C, the change is from 18 nm after 30 min to 42 nm after 16 h. There must also be additional thickening behind the growth front to account for the variation of melting point with radial distance.

As a consequence of isothermal thickening, the density of branches at fold surfaces may increase somewhat, although it will already be high given that the initial

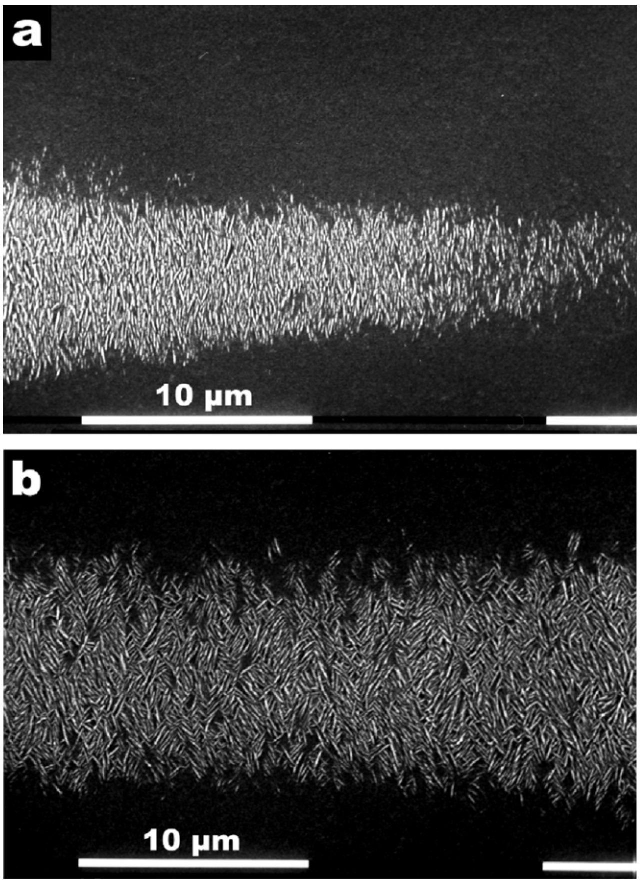

Fig. 7. Radial views of row structure morphology at 124 °C after (a) 4.5; (b) 15 h.

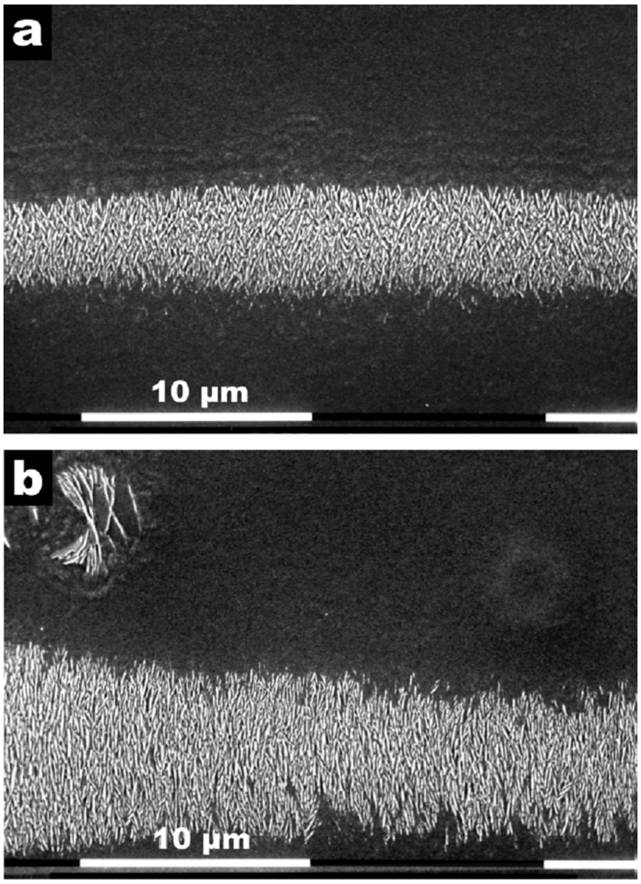

Fig. 8. Radial views of row structure morphology at 125 °C after (a) 12; (b) 25 h.

lamellar thickness is about equal to the average interval between branches, with fractional crystallization having rejected all sequences shorter than the initial fold length. Thickening will tend to bury branches further into surface regions, introducing what we term interstitial surface defects or ISDs. A buried branch would be surrounded by longer stems, although they must be fewer than the number of folds otherwise they could not be buried. The compressive stress in fold surfaces will, therefore, increase with thickening, predominantly from the latter cause. In this condition it is possible for a lamella spontaneously to reduce its free energy in three ways. First the inclination of fold-surface normals to the chain axis could rise to increase the surface area per fold stem. Second, following recent work on monodisperse branched alkanes [8], it could bend with ISDs located in the convex surface. Third it could twist helicoidally [9] partly to relieve the surface stress in those portions which it extends.

There is no strong evidence that the first possibility occurs, there being no evidence that isothermal thickening produces a higher fold-surface inclination than ~{201} in the branched polymer. But it cannot be excluded because the maximum measured inclination is ~35°, a little more than the ± ~ 30° observed in e.g. Fig. 4(a) and fracture surfaces, a figure on the low side of published distributions for ~{201} fold surfaces in linear polyethylene [6]. The second possibility is a new factor revealed by monodisperse alkanes with a single central branch which, at high crystallization temperatures, form lamellae rolled into cylinders of uniform diameter ~1 μm but decreasing for a longer branch [8]. The molecules have a hairpin conformation and all branches lie in the outer, convex fold-surface in a habit, which, forming spontaneously must be one of lower free energy. It is of particular relevance to the present paper because ISDs would tend to move to the convex fold-surface thereby introducing a bend component of stress. They are able to do so because of the ease with which stems can translate through a lamella so that a branch at a fold-surface can readily be converted into an ISD and vice versa. Bending, once begun, will tend to increase, provided there is no physical bar to increased curvature, leaving branches in the concave surface and ISDs in the convex. The implication is that the presence of ISDs makes a planar lamella unstable against bending.

The role of the third factor is unclear. There is no evidence for simple helicoidal twisting in linear polyethylene but it has been pointed out that the concentration of stress along the turning points of an S-profiled lamella is a favourable one for reducing elastic energy by twisting [10].

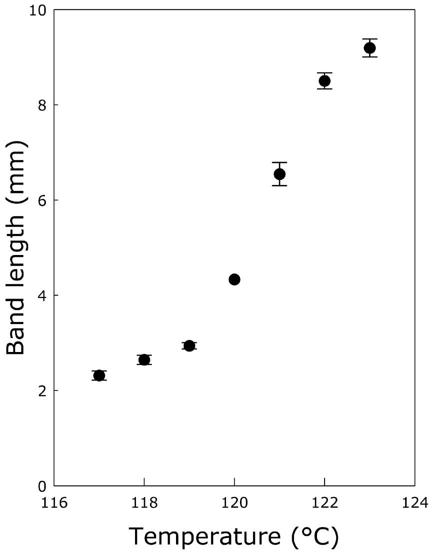

Fig. 9. Plot of band interval in row structures, from the fibre to the first dark band in the polarizing microscope plotted against crystallization temperature.

To address the question of why late banding occurs for slow growth of linear-low-density but not linear polyethylene, one notes that substantial isothermal thickening does not lead to twisting growth in the linear polymer and would be expected, of itself, to maintain an ordered fold-surface with its lower free energy. The irregularly placed ISDs produced in the branched polymer would be analogous to as-grown folds in faster growth being randomly, not crystallographically, placed. As such they could, in principle, parallel behaviour for linear polyethylene and give the oldest, central part of a lamella a shallow S-profile inclined to **b**, the growth axis provided the fold-surface inclination is increased over the initial value.

Phenomenologically there is a great similarity between banding for slow and fast growth, both having planar lamellae at the growth front with S-profiles developing behind notwithstanding the difference in the initial internal molecular orientation, inclined and perpendicular, respectively. The shallow S-profiles superposed on the initial inclined habit with a small reduction in fold-surface inclination at the edges are strong evidence for a transverse bending moment arising from an asymmetric disposition of ISDs. This asymmetric disposition also implies that the axis of the S will be inclined to the **b** axis. Once an S-profile is present in the older part of a lamella, it would generate a twisting stress ahead of it probably leading, as in the linear polymer, to isochiral giant screw dislocations, placed to one side of a lamella at each of which the twist would be amplified and an effective increment of twist inserted [10].

The different magnitudes of the stresses causing banding for faster and slower growth, corresponding to irregularly placed folds and ISDs, respectively, are revealed in Fig. 9. This plot of band length, i.e. interval to the first dark band from the fibre surface, as a function of crystallization temperature increases its gradient when faster gives way to slower growth with a corresponding increase in band length. The gradient more than doubles (from 0.9 to 2.1 μm K^{-1}) as crystallization temperatures rises through 119 °C revealing a change to less twisting. From 119 to 120 °C there is a large change in band length, from 2.9 to 4.3 μm. As there will be only a small change of lamellar thickness, it follows that it is the twisting stress which is reduced when fold surfaces are initially ordered in line with the lower concentration of defects than folds.

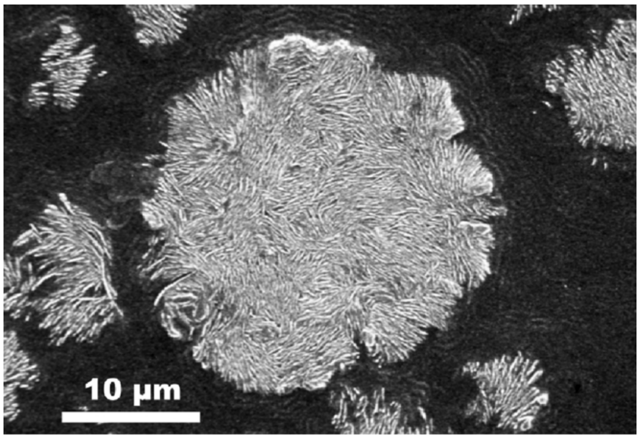

Fig. 10. Cellulation in a spherulite, adjacent to a row, grown at 123 °C for 18 h.

With the change to twisted growth, the pattern of cellulation has to change from that appropriate to lamellae growing perpendicular to the fibre axis. The second kink shown in Fig. 3 for 123 °C shows that the segregant concentration at the growth interface is once again restored more or less to zero with only a comparatively gentle decrease in growth rate thereafter. As Fig. 6 makes clear, growth then proceeds in cells a few microns across, the dimension increasing with crystallization time, within each of which lamellar groups twist as a whole but not necessarily in the same sense as their neighbours. This is a possible reason for the presence of the 'reverse S' profiles rarely if ever observed in linear polyethylene which may be the lamellae which had begun to adopt the opposite chirality to the majority but had to twist in the opposite sense within the bundle. This does not affect the dark band in the polarizing microscope as lamellar planes coincide (Fig. 6(c)) for twists of ±90° but the cells stay distinguishable and separate. A somewhat similar pattern of cellulation occurs in adjacent spherulites (Fig. 10) in which there are individual units but with no organized banding evident.

5. Conclusions

The principal conclusions of this work are, first, that the rate of radial advance of crystallizing row structures of linear-low-density polyethylene slows continually as the segregation of more branched sequences or molecules proceeds until it is temporarily restored more or less to the original value at the onset of cellulation. The pattern of cellulation changes, with a further kink in the rate plots, when growth becomes banded.

This is the first observation of banding for slower growth in polyethylene, i.e. when fold surfaces are initially ordered. It is a consequence of surface stress created when isothermal lamellar thickening brings branches into fold surfaces in contrast to the usual situation, for faster growth, when banding relieves the stress of the initial fold-surface packing.

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