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On pulsating growth rates in banded crystallization of polyethylene

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Abstract

Three polyethylenes, two Ziegler–Natta linear-low-density materials and one linear polymer, have been crystallized on fibres as linear nuclei to give banded row structures. Whereas the radii of rows of the linear polymer increase linearly with time, those of the two branched polymers do not do so but oscillate around the linear plot in phase with the bands; maximum growth rates occur at the start of both flat-on and edge-on bands. The phenomenon is related to segregation of more-branched molecules which depresses the growth rate. Accelerated growth occurs at the start of the respective bands when lamellae no longer grow parallel but diverge. For edge-on bands this occurs by point-like nucleation and cellulation. Diverging lamellae grow faster than a parallel array because of lower segregant concentration. Accelerated growth may also be correlated with branching because asymmetric branching, with its accompanying increment of lamellar twist, will allow the growth front systematically to reach regions of reduced segregant concentration. A small but repeatable effect in the linear polymer, for which segregation is insignificant and kinetics are not slowed, raises the wider question of whether branching affects growth rate in general.
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Keywords: Branched polyethylene; Growth kinetics; Banded row structures

1. Introduction

The well known observation that the isothermal radial growth rate of polymer spherulites is constant, e.g. [1], depends upon steady-state conditions pertaining at the crystal/melt interface. This is not immediately the case when substantial segregation of molecular species lowers both local equilibrium melting point and isothermal supercooling and a steady state has then to be established. In such circumstances recent work, initially for linear-low-density polyethylenes crystallized on linear nuclei then for spherulites under conditions of high segregation and cellulation, has demonstrated continuous declines in growth rate with time and radial distance [2,3] to asymptotic limits [4,5]. Contrary to this general trend, an increase in growth rate was observed in one circumstance, Fig. 3a in Ref. [2], namely when lamellae growing in a row structure began to grow parallel to the row instead of perpendicular to it. We now report extensive observations which generalize this first observation and show that, in linear-low-density polyethylenes, the growth rate of banded row structures not only increases with a change to flat-on lamellar orientation but also oscillates in phase with the bands. Increases are, however, found at the onset of edge-on as well as flat-on bands which requires development of the above explanation into a more general form. This is suggested by the morphology which shows that the acceleration of growth at the start of an edge-on band is linked to point-like nucleation of cellular growth at the instantaneous interface instead of propagation of a lamellar uniform array. This change is favoured because it is faster, there being less consequent segregation. It may also be that branching promotes faster growth as, being asymmetric with an associated increment of twist, it will give systematic access to regions of reduced segregant concentration. A small but repeatable pulsation superposed on the growth of linear polyethylene, for which the influence of segregation has to be excluded, raises the question of whether branching affects growth rate more generally, as has also been recently observed in different circumstances for the long *n*-alkanes [6,7].

2. Materials and experimental

One linear and two linear-low-density polyethylenes have been used in this work. The linear material was Sclair 2907 (du Pont, Canada) for which $M_w = 104,000$; $M_n = 11,300$. The two branched polymers have been studied previously

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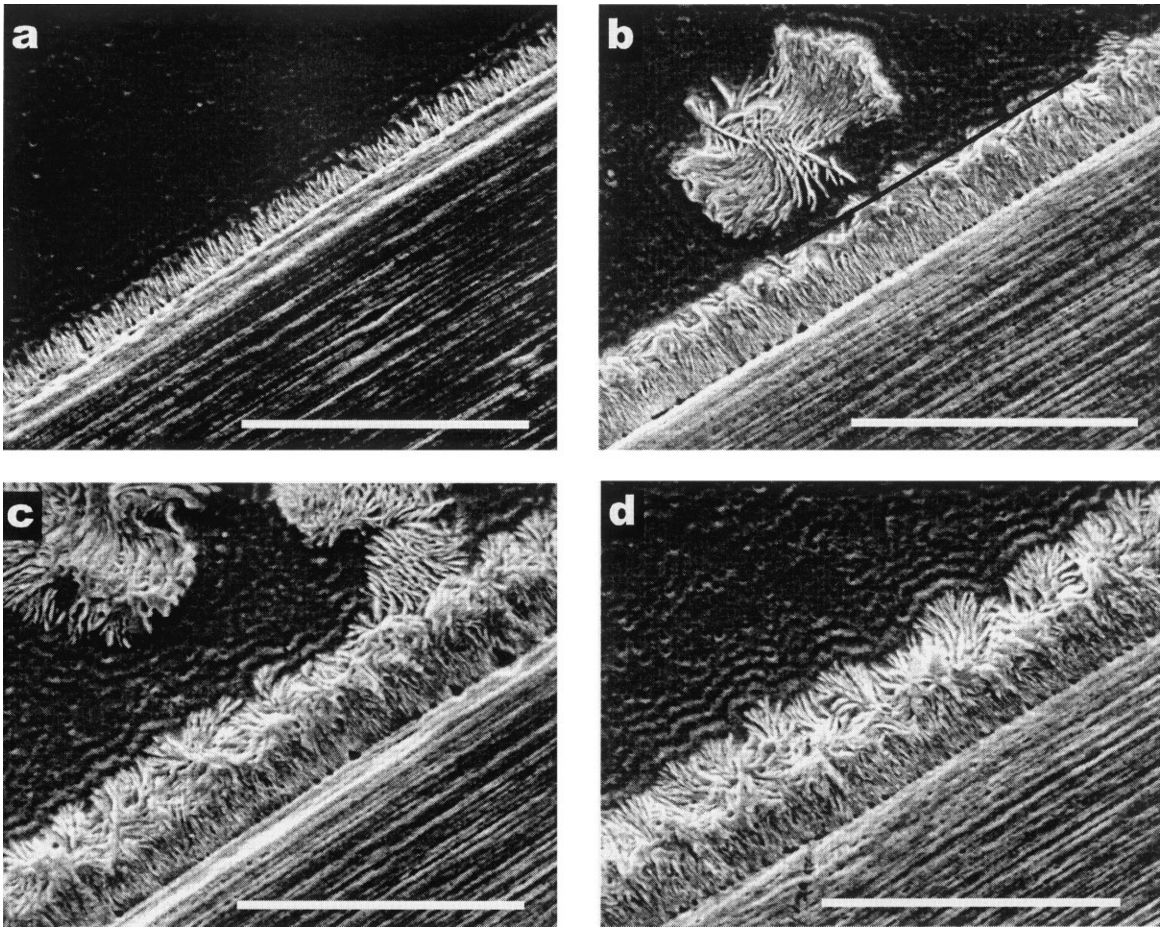

Fig. 1. (a) The uniform, fine initial structure of a row in polymer B (within a quenched matrix of banded spherulites) with lamellae perpendicular to the fibre axis and sharing its *c*-axis; 3 min crystallization at 121°C. Bar = 10 μm. (b) With increasing diameter, some flat-on lamellae have developed at the outside of a row of polymer B which are more or less parallel to the fibre axis; 7 min crystallization at 121°C. Note that radius of the adjacent spherulite is wider than that of the row indicating its faster growth rate due to a lesser segregant concentration. Bar = 10 μm. (c) An early stage in the development of the second edge-on band in a row of polymer B; 9 min crystallization at 121°C. Bar = 10 μm. (d) The second edge-on band can be seen apparently to have grown out from points (in projection); 11 min crystallization at 121°C. Bar = 10 μm.

by us [8]; they are again denoted B and C. Both were synthesized by Ziegler–Natta catalysis and were supplied by Dr P. Jääskeläinen of Borealis Oy, Finland. Polymer B has 9.7 butyl branches per 1000 carbon atoms and average molecular weights: $M_w = 121,000$; $M_n = 27,000$ while polymer C has 15 butyl branches per 1000 carbon atoms; $M_w = 140,000$; $M_n = 30,000$.

The primary observation made was of row diameter as a function of isothermal crystallization time. To this end, specimens containing high-melting polyethylene fibres as linear nuclei were prepared as follows. Firstly, a flat disc of the chosen polymer, $\sim 20 \times 30$ mm^2 across and ~1 mm thick, was fabricated from the appropriate quantity 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. This disc was then melted, for 3 h at 200°C in a Mettler FP82HT hot stage with nitrogen flow, a procedure designed to reduce the number of very long molecules in the sample and hence the density of spherulitic nucleation while leaving the remaining molecular distribution essentially unaltered [9–11]. As a consequence, crystalline entities grow larger before impinging which in turn enables the growth length to be measured over a wider range. At the end of this time the 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 size which were stored in labelled jars ready to prepare a fibre/polymer composite as below.

A specimen containing about seven highly oriented Tekmilon™ polyethylene fibres (Mitsui) within a polyethylene matrix was prepared, between the microscope slide and the cover slip. Initially the fibres were stacked parallel on a microscope slide with their ends lightly adhered with Araldite™. This slide was inverted and placed carefully on one of the stored square discs already sitting on a cover slip at 130°C on the Kofler hot bench, applying light pressure to the already-melted disc. The resulting fibre/polymer

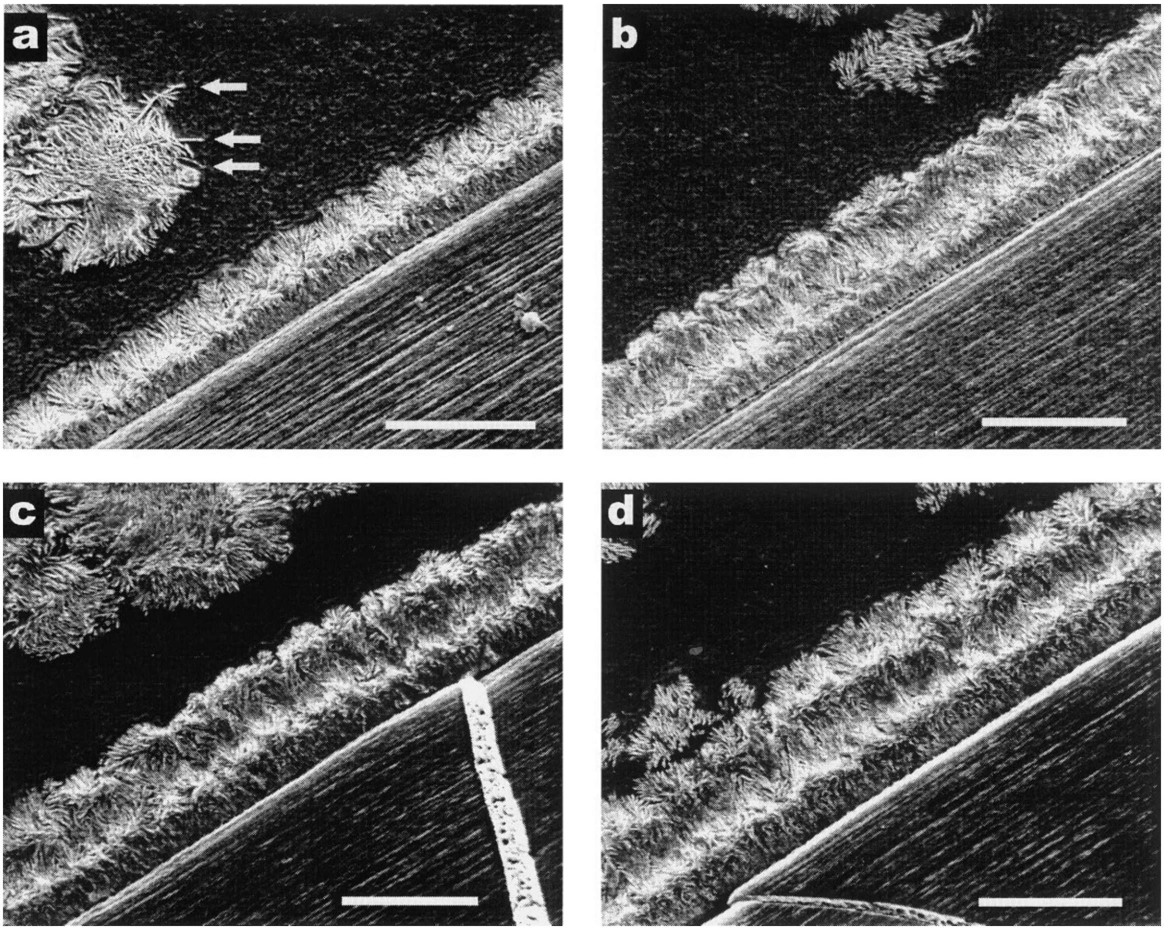

Fig. 2. (a) A nearly uniform, but somewhat coarsened, second edge-on band at the outside of a row in polymer B. Note also the wide separation of dominant lamellae in the adjacent spherulite, three of which are indicated by arrows. 13 min crystallization at 121°C. Bar = 10 μm. (b) Development of the second flat-on band in a row of polymer B; 20 min crystallization at 121°C. Bar = 10 μm. (c) The start of the third edge-on band; 22 min crystallization at 121°C. Bar = 10 μm. (d) A nearly complete third edge-on band in which there is again evidence for apparent point-like nucleation.; 24 min crystallization at 121°C. Bar = 10 μm.

composite, with microscope slide and cover slip, was then transferred to the Mettler hot stage at 130°C for 5 min before the temperature was lowered to the required crystallization temperature using a Mettler FP90 central processor. The composite specimen was quenched in an ice–water mixture after the selected time of crystallization and carefully removed from the slide and cover slip. Nomarski differential interference microscopy showed that the fibres had become embedded with their centres ~60 μm below the surface of the disc.

To reveal the embedded fibres and measure the diameters of the row structures grown on them, all specimens were etched for 4 h 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. The etching time and etchant formulation were selected so that the final plane surface of each specimen passed close to the fibre axes i.e. exposing a diametral section. This was readily checked with the Nomarski microscope which showed the width of fibres to be close to their nominal diameter of 40 μm. Such etched specimens were coated with gold prior to examination in the scanning electron microscope, SEM. Quantitative measurements of row diameter, for all etched specimens, were obtained using the SEM, making sure that any non-diametrical sections, i.e. with a fibre width markedly less than the nominal diameter, were excluded.

The numerical data for each point in the figures, with associated statistical parameters, were obtained from three to five SEM photographs per sample, taking two or three measurements of growth length per photograph, i.e. around 10 measurements were taken for each data point. The growth length was measured directly on each photograph, from the fibre surface to a line tangential to the edge of the structure and parallel to the fibre axis as shown in Fig. 1b.

When it was desired to view the rows down the growth direction, a similar procedure was used but with a shorter etching time. Observations of morphology in planes normal to the nucleating fibre were made on specimens cut open with a microtome prior to etching.

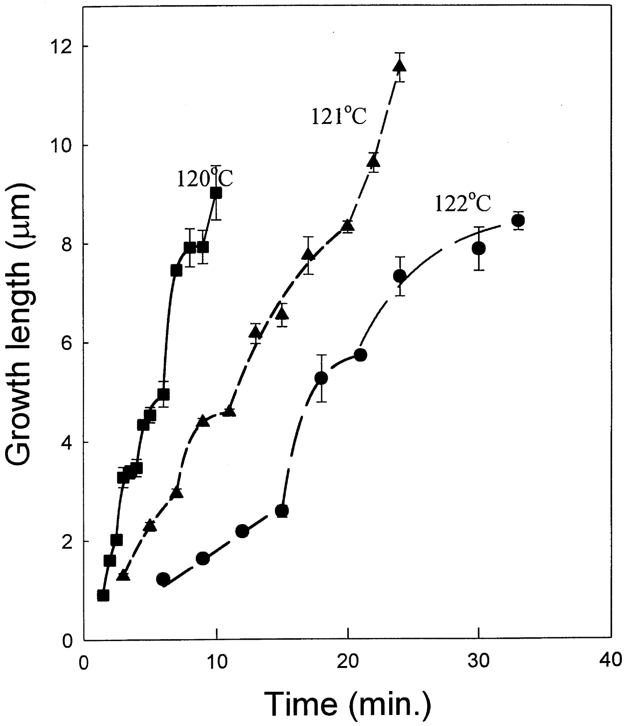

Fig. 3. The average radial growth length of row structures of polymer B against crystallization time for 120, 121 and 122°C.

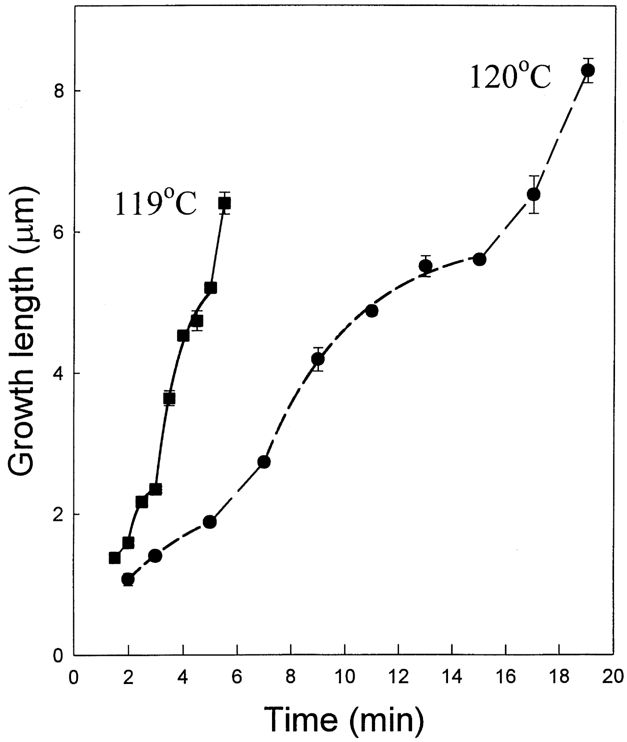

Fig. 4. The average radial growth length of row structures of polymer C against crystallization time for 119 and 120°C.

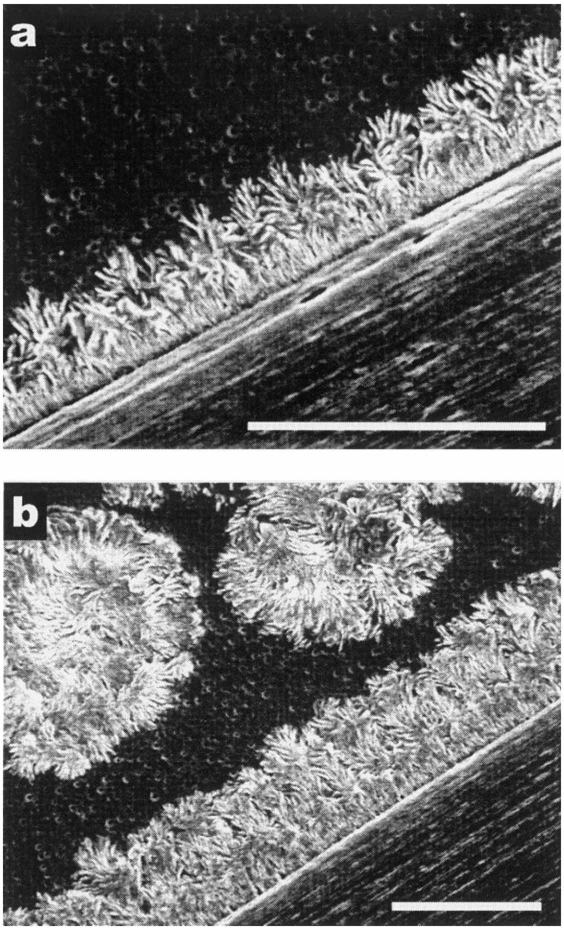

Fig. 5. (a) A banded row structure of polymer C which has grown out to the second edge-on band; 9 min crystallization at 120°C. Note that by comparison with polymer B in Fig. 1d, the texture of this more-branched polymer is less uniform. Bar = 10 μm. (b) Further development of a banded row in polymer C out to the third edge-on band; 19 min crystallization at 120°C. Bar = 10 μm.

3. Results

All micrographs in the figures are SEM images of etched surfaces. Figs. 1 and 2 show the morphologies of different rows prepared for increasing crystallization times at 121°C. Fig. 1 covers the development of the first band, i.e. the first 180° rotation of lamellar orientation, Fig. 2 that of the second band. In Fig. 1a the first micron of the row structure, grown in 3 min at 121°C, has lamellae growing perpendicular to the fibre axis, sharing its *c*-axis direction but there is already some coarsening, i.e. fewer lamellae per unit length of row than initially, over this interval which may be facilitated by the inability of those lamellae nucleated, but not orientated with their *b*-axis in the plane of the paper, to propagate radially. Fig. 1b–d show the sequence for 7, 9 and 11 min crystallization, respectively, through the first band of flat-on lamellae to the second edge-on region, i.e. a 180° rotation from the original. The line in Fig. 1b is that used for measurement of the row length, ignoring the local protuberances due to the developing cellulation. Notice also

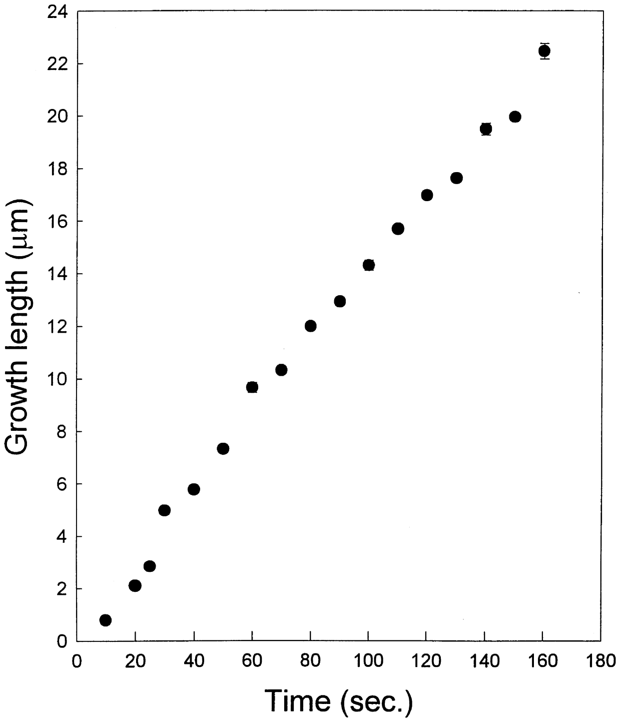

Fig. 6. The average radial growth length of rows of Sclair 2907 crystallized at 123°C.

in this figure that the adjacent spherulite is much wider than twice the row, showing clearly that spherulites, with their more open texture, grow faster. This important and central point is reinforced by the microstructure of the spherulite nearest the row in Fig. 2a in which dominant lamellae (three of which are arrowed) are separated by microns, dimensions which will reduce the local segregant concentration well below that pertaining to the constrained growth in the row. Fig. 2a–d are for rows crystallized at 121°C for 13, 20, 22 and 24 min, respectively, which cover the development of the second 180° rotation from the original.

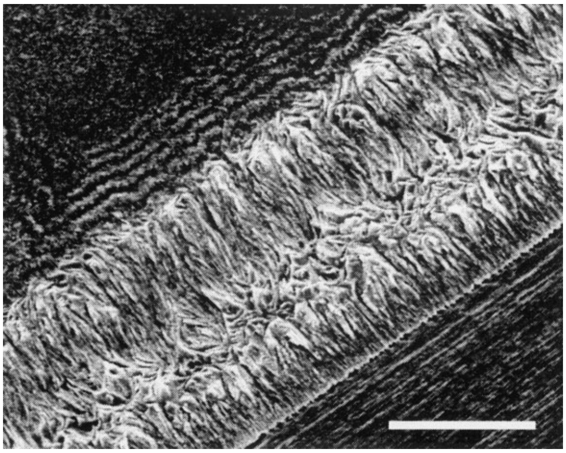

Fig. 7. Uniform banding in a row of Sclair 2907; 130 s crystallization at 123°C. Bar = 10 μm.

When length data measured from these and equivalent micrographs are plotted against time they appear as shown in Fig. 3, together with corresponding data for crystallization at 120 and 122°C. Curves have been drawn through the points to show the repeated pulsations of radial length and thus growth rate. Comparison with the morphologies of Figs. 1b and 2b shows that these are in phase with the banding, as is discussed in detail below.

Polymer C behaves in a similar way while also exhibiting the greater degree of cellulation expected of a more-branched polymer. Fig. 4 displays length data for 119 and 120°C while illustrations of rows grown at 120°C for 9 and 19 min, through the first and second bands, are in Fig. 5a and b, respectively. The latter also shows clear cellulation at the periphery of the adjacent spherulites as well as in the rows.

When the same methodology is applied to banded rows of the linear polyethylene Sclair 2907 grown at 123°C, the length data (Fig. 6) are essentially linear with time, with just a trace of systematic departures, whose significance is discussed in the next section. Figs. 7 and 8 show the uniform banding for this material and crystallization temperature from two different perspectives. Fig. 7 has the plane of the row in the paper while Fig. 8 shows an oblique view of a plane normal to the nucleating fibre. The latter also brings out the different etching responses of the fibre and lamellae crystallized on it which are, in order of decreasing resistance, the central fibre, flat-on lamellae then edge-on lamellae. The craters on the etched fibre reflect internal lateral ordering as discussed elsewhere in relation to a new model of fibre structure [12,13].

Consideration has so far been restricted to lamellae either perpendicular or parallel to the row in which two special orientations there is no inherent restriction on lamellar width. It is, however, impossible to rotate one of these orientations continuously into the other so that the geometry of banding requires lamellar widths to be finite. Relevant information comes from viewing the rows in other perspectives. Fig. 8 complements Fig. 7 in showing the circular symmetry with lamellae initially in this plane and perpendicular to the row. In Fig. 9 are views looking down the growth direction at different depths which reveal widths of lamellae and their lateral ordering directly. Fig. 9a is close to the fibre with lamellae still planar and perpendicular to its length. At greater distance (Fig. 9b) the pattern of S-profiled dominant lamellae and infilling subsidiaries characteristic of banded polyethylene spherulites [14] is present as Fig. 9c identifies in detail. The transformation of one type of lamella to the other is central to the development of banding as will be discussed in a forthcoming publication.

4. Discussion

The constancy of the isothermal growth rate of spherulites is one of the most familiar features of polymeric

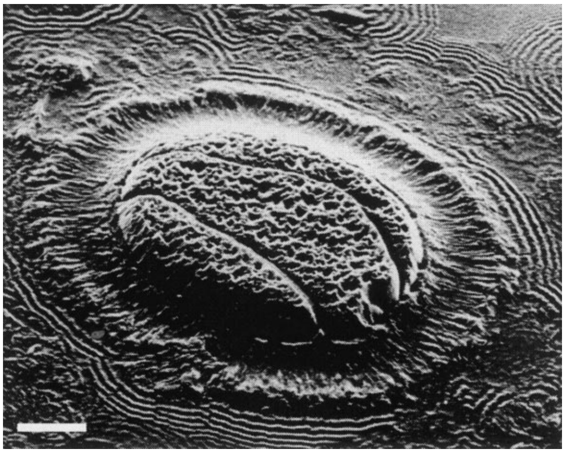

Fig. 8. An etched equatorial section normal to a row structure of Sclair 2907, viewed obliquely; 90 s crystallization at 123°C. Bar = 10 μm.

crystallization and normally extends to growth nucleated on lines or surfaces to form row structures [15,16] and transcrystalline layers, respectively, [16,17]. The implication is that conditions at the crystal/melt interface remain constant as growth proceeds. But this will no longer be the case at the start of growth when the local molecular constitution of the melt is changed by rejection, segregation and accumulation of certain species at the growth front. When linear-low-density polyethylenes crystallize, the more-branched molecular sequences are so rejected leading to a progressive lowering of the local equilibrium melting temperature, isothermal supercooling and growth rate [4,5]. In the investigation of such phenomena, row structures offer two main advantages over spherulites: they maximize segregant concentration and allow changes of the pattern of crystallization to be more readily recognized as departures from a uniform array; moreover, crystallographic directions are identifiable by inspection.

In the ensuing discussion we shall first establish the link of increases in growth length and rate with the start of a new band, when lamellae are either edge-on or flat-on, i.e. perpendicular and parallel, respectively, to the linear nucleus. The two orientations are distinct in row structures but would be identical in spherulites nucleated from a point. The second item to note is that such increases accompany diverging lamellar growth which, at least for edge-on bands, is accomplished by point-like nucleation, in projection, as opposed to propagation of a uniform lamellar array (Fig. 10). Diverging growth is faster than that of parallel lamellae because with fewer and more-separated lamellae it gives less segregation and lower segregant concentrations but growth will slow as diverging lamellae become constrained by cell boundaries and bend to grow normal to the row. Thirdly we note that asymmetric lamellar branching provides a possible systematic and additional means of reaching regions of lower segregant concentration giving faster growth. Finally, the question is raised whether branching, in general, may temporarily affect growth rate

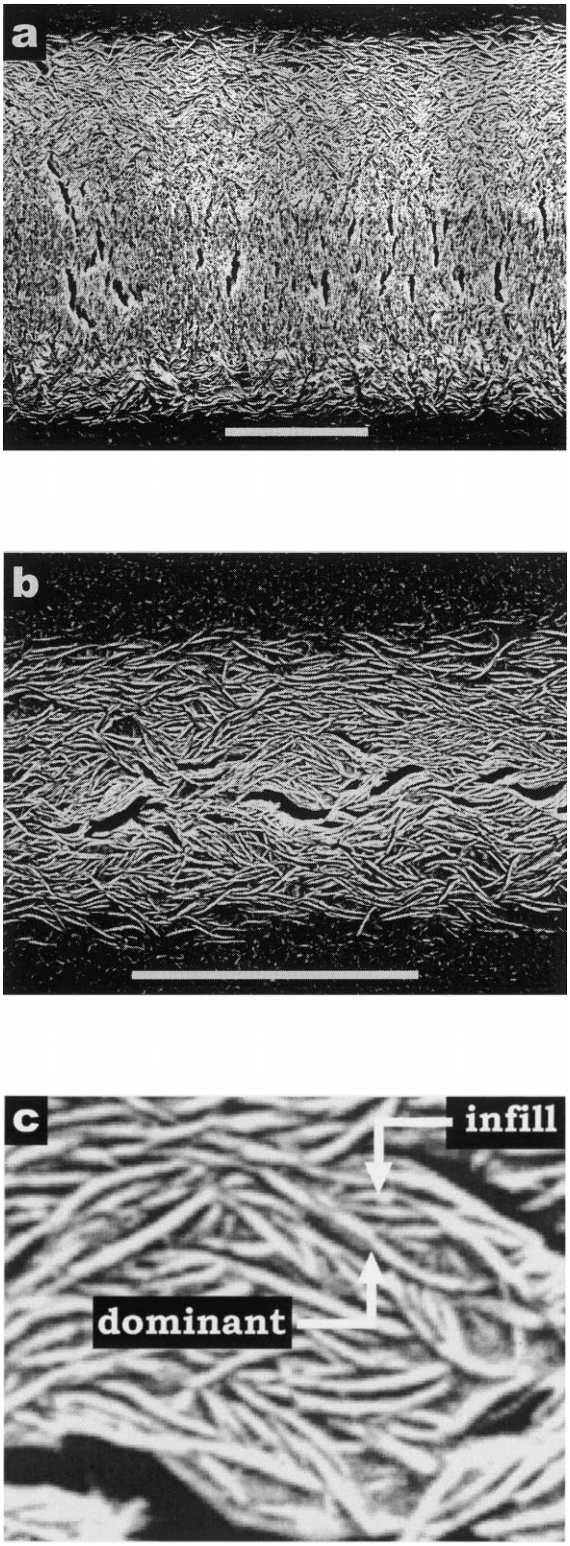

Fig. 9. (a) Lamellar detail in a plane parallel and close to the nucleating fibre's surface in a banded row structure of Sclair 2907; 40 s crystallization at 123°C. Note that lamellae are initially perpendicular to the length of the fibre. Bar = 10 μm. (b) A similar view to Fig. 9a but of a plane farther away from the nucleating fibre; 40 s crystallization at 123°C. Note that S-profiled lamellae have now developed whose widths are more or less parallel to the fibre axis. Bar = 10 μm. (c) Detail of the centre of Fig. 9b identifying dominant and infilling lamellae.

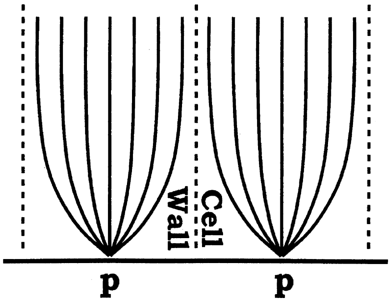

Fig. 10. Schematic illustration of point-like nucleation in projection at points p and the inferred location of intervening cell walls.

in the absence of segregation and cellulation, complementing measurements on the long-chain *n*-alkanes [6,7].

The pulsations of growth rate reported in this paper are primarily a consequence of segregation, being essentially absent from the linear polymer. As increasing segregant concentration reduces the growth rate the likely explanation is that faster growth occurs in response to reduced segregant concentration at growing lamellar interfaces. It has previously been pointed out, in relation to a single faster-growing entity within a row (Fig. 3a of [2]) that segregant concentration will be less, and growth rates correspondingly higher, when lamellae lie parallel to the row than when they are perpendicular to it. The cause is geometrical because, in the former case, lamellae are able to hinge around the nucleus and grow radially to it, increasing their separation and reducing segregant concentration as they advance. This contrasts with the initial situation when, growing perpendicular to the row, lamellae are close-packed. Then, their separation can only increase if some are eliminated—as may be seen happening in Fig. 1a—coarsening the texture. But coarsening accompanies a continual fall in growth rate, because of continued segregation, with both tending to asymptotic limits [5]. The rotation of lamellae to a flat-on orientation in banding changes matters allowing growth to accelerate towards the faster speeds found in the radial growth of spherulites.

While this suggestion is sufficient to account for faster growth of flat-on lamellae within edge-on surroundings in the absence of banding [2] it would predict just one increase per cycle when banding is present. Fig. 3 shows that, in practice, there are two increases, not only at the start of the flat-on lamellar orientation as expected on the above argument but also at the start of the edge-on region.

Examination of the relevant morphologies shows that the kink at 3 μm in the growth length of Polymer B at 120°C (Fig. 3) may be seen in Fig. 1d to correspond to the onset of point-like nucleation, in projection, of lamellae growing in edge-on orientation, i.e. effectively in cells. This is also the case for the kink at 8.3 μm (Fig. 2d). On the other hand, the kink at 4.6 μm corresponds to the onset of flat-on lamellae (Fig. 2b). A similar situation pertains for Polymer C. Fig. 5a reveals that 2.7 μm, the location of a kink in Fig. 4 for growth at 120°C, marks the onset of edge-on growth nucleated at points. The further kink in Fig. 4, at 5.6 μm, is when the flat-on portion of the band begins (Fig. 5b).

It is evident that the pulsating growth involves repetitive point nucleation, in projection, of edge-on bands at sequential growth fronts. Lamellae diverge from these points thereby reproducing the geometry cited before for flat-on bands but in a different context. Diverging geometries may thus be expected to commence at the start of both edge-on and flat-on bands, although the latter would be invisible from the perspective used. Such divergence, either from apparent point nucleation or from geometrical hinging, represents a faster-growing mechanism, because of lower segregant concentration, than an array of parallel lamellae.

Similar ‘point nucleation’ was observed at the instantaneous edge of row structures previously [2] and then suggested to arise, when growth had slowed sufficiently, from lateral fluctuations in segregant concentration along the growth front, occurring where this was least. There is support for this view here in the fewer lamellae located to the side of the point nucleus presumably reflecting high concentrations of uncrystallized rejected species in these sites. The ‘points’ divide the growth front into cells which will, so long as divergence persists, grow faster than a parallel array. However, when lamellae meet at cell boundaries those continuing to grow are constrained to bend and form a parallel array whose growth rate will be correspondingly less because of higher segregant concentration.

The question which then arises is why should ‘point nucleation’ in rows occur systematically at the start of edge-on bands. The answer may be that this location is singular in that it is where, if they were continuous, lamellae would have to start to grow parallel again after a flat-on band in which they could diverge radially. The restoration of a parallel array, probably with a reduced number of lamellae, would be an impediment to growth. This location would, therefore, be that of maximum difficulty in maintaining growth and lamellar continuity, making it particularly vulnerable to the onset of growth able to start at just one site. The significance of the ‘point nucleation’ would, accordingly, be that this offers the system the fastest way ahead.

A generalization of this concept may contribute to the acceleration at the start of a flat-on band. It is probable that a uniform array of lamellae will be vulnerable to being replaced by others growing faster due to local secondary nucleation whenever, to maintain the array, all lamellae would have to perform identical transformations, notably branching which rotates lamellar planes, in synchronism. Any one lamella able to fluctuate ahead of its neighbours would gain a competitive advantage in growth.

An additional factor to consider is that low segregant concentrations may be reached systematically in twisting morphologies when a lamella branches. This is because the uniform macroscopic twist around the radius is not found in individual lamellae. For them, twist occurs mostly in increments associated with screw dislocations [18,19]. Moreover, these screw dislocations are asymmetrically distributed [20]. This would have the important consequence that a change of azimuth around the radial growth direction would enable growth to move into regions of lower segregant concentration than would otherwise be the case. In consequence, one would expect branching and acceleration to be correlated when there is sufficient segregation.

It is too soon to draw detailed conclusions of whether and, if so to what extent, these observations on rows, particularly the systematically repeating 'point nucleation' under cellulating conditions, may be relevant to the banded crystallization of spherulites undergoing segregation. As pointed out earlier, rows and spherulites differ in that the former alone have a frame of reference which distinguishes between planes parallel and perpendicular to the nucleus. Nevertheless, there are similarities in behaviour, beginning with cellulation. One of us [21] has observed that in such circumstances, spherulites of linear-low-density polyethylene grow by leading lamellae slowing and being overtaken by others from the rear which avoid high concentrations of segregant, so that the growth front advances unevenly in space and time. Moreover, unpublished direct measurements of spherulites with large band period growing in highly branched polyethylene have also shown them to advance in an oscillatory manner.

In conclusion we consider the case of the linear polyethylene, Sclair 2907, which is intriguing in that comparison of Figs. 6 and 7 shows that the small deviations from linearity, at 5 and 10 μm, are at the onset of the flat-on and edge-on zones, respectively, i.e. paralleling the behaviour of their branched counterparts but to a much smaller extent. Unlike the latter, however, our measured growth kinetics do not decrease with distance (at 127, 128 and 129°C) even when there is cellulation. As previous work [22] has shown a small degree of coarsening of texture for crystallization at 127.5°C, indicative of a little fractionation of shorter molecules, a process which increases with further rise of crystallization temperature, it would have been possible to claim, a priori, that a small amount of segregation, of shorter molecules, might occur at 123°C with some reduction of isothermal supercooling and growth rate. This possibility is negated by the maintained linearity of kinetics with radial distance under higher degrees of segregation cited above. Nevertheless, results similar to Fig. 6 in which a small systematic perturbation is superposed on the linear behaviour, have been found by us in repeated experiments at both 123 and 124°C, providing grounds for considering this small effect in linear polyethylene to be real and unrelated to segregation. This brings the intriguing possibility that kinetics may be directly affected by branching itself, as has been found recently for extended-chain crystallization of monodisperse long *n*-alkanes albeit in a different context and with opposite sign [6,7]. In this instance the most likely candidate would be simplification of interfacial condition with change of azimuth when branching occurs. At present, however, this can only be a basis for discussion.

5. Conclusions

The principal findings of this work are

(1) The growth rate of banded rows of linear-low-density polyethylene pulsates in phase with the banding, increasing at the start of each edge-on and flat-on band.

(2) The acceleration is linked to the onset of diverging as opposed to parallel growth of lamellae. In the case of edge-on bands this is a result of repetitive 'point nucleation' at their start. Under segregating conditions, when growth rate is depressed, diverging growth is faster than the propagation of a uniform lamellar array.

(3) Asymmetric branching, with its accompanying increment of lamellar twist, offers an additional and systematic means of reducing local segregant concentration at the growth front and achieving faster growth.

(4) Small but repeatable pulses in linear polyethylene, when kinetics are not slowed by segregation, raise the issue of whether branching, with its associated increment of twist, may directly affect the growth rate in the absence of segregation and cellulation.

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