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On deformation mechanisms of β-polypropylene 3. Lamella structures after necking and cold drawing

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Abstract

A β-PP sample was stretched to necking at room temperature and the morphologies of the deformed material over the necking region were examined with SEM and TEM. Before yielding, the strain was mainly accommodated between horizontal lamellae. Nevertheless, some vertical lamellae were stretched to break when the material approached yielding. At the yield point, many melt spots and deformation bands across vertical lamellae emerged. Some vertical lamellae became highly stretched and their thickness reduced significantly while the neighboring horizontal lamellae rotated towards the loading direction. At the upper shoulder of neck, the melt spots near the poles of spherulites enlarged and elongated in the stress direction, meanwhile, strain-induced crystals appeared in the melt spots. Furthermore, many vertical lamellae were broken into short fragments and some horizontal lamellae were sheared in the loading direction when the adjacent vertical lamellae were stretched heavily. In the tapered section of the neck, the original β-spherulites were shattered due to excessive crazing and deformation bands and some highly drawn material domains were formed. In turn, more shear bands were generated roughly along the loading direction. The horizontal lamella fragments rotated towards the loading direction and fed into the material flow continually. Finally, the lamella–spherulite structure was converted into an oriented fibril structure in the cold drawn material. Based on the observed results the deformation mechanisms of necking and cold drawing were discussed.

Keywords: β-Polypropylene; Necking and cold drawing; Deformation mechanism

1. Introduction

The unique feature of crystalline plastics, cold drawing following necking, has been observed for a long time and it has also been recognized that the spherulite structure in the undeformed sample converts into an oriented fibrous structure in the cold drawn material. However, how the spherulite converts into fibrous structure during cold drawing is still not fully clarified in the literature. Peterson [1] and Bowden and Young [2] first suggested that the movement of screw dislocation along the chain direction was operable in polyethylene (PE) under the action of stress and responsible for plastic deformation. Peterlin et al. [3,4] noticed some crystal blocks which were separated with amorphous layers in the drawn single crystal PE mats and extremely thin PE film. These blocks have a similar dimension as the thickness of the single crystals before deformation and were believed to be the fragments of the precursor crystals. Hence, Peterlin proposed [5] that the initial lamellae were broken into crystal blocks and reorganized into microfibrils during cold drawing. Flory and Yoon [6] inferred that the neighboring lamellae in melt-crystallized polymers should be profusely interconnected as a random switchboard. The large irreversible deformation in semicrystalline polymers, such as occurs in cold drawing, has to involve destruction of pre-existing crystalline regions followed by recrystallization in melt phase.

Indeed, when PE was stretched above 70°C some continuous crystalline fibrils up to several μm long were observed in the drawn material [7,8]. This was thought to be a consequence of stretching induced melting followed by crystallization at the stretching temperature. The same conclusion of stretching induced melting of PE was also derived from the results of small angle neutron scattering [9]. Furthermore, a recent investigation [10] of the contents of β-phase in a β-PP sample over the necking region demonstrated that the β-phase could be stretched to melting locally at room temperature and then recrystallize into α-phase. However, how the β-spherulite converted to α-fibrils during necking has not been reported yet.

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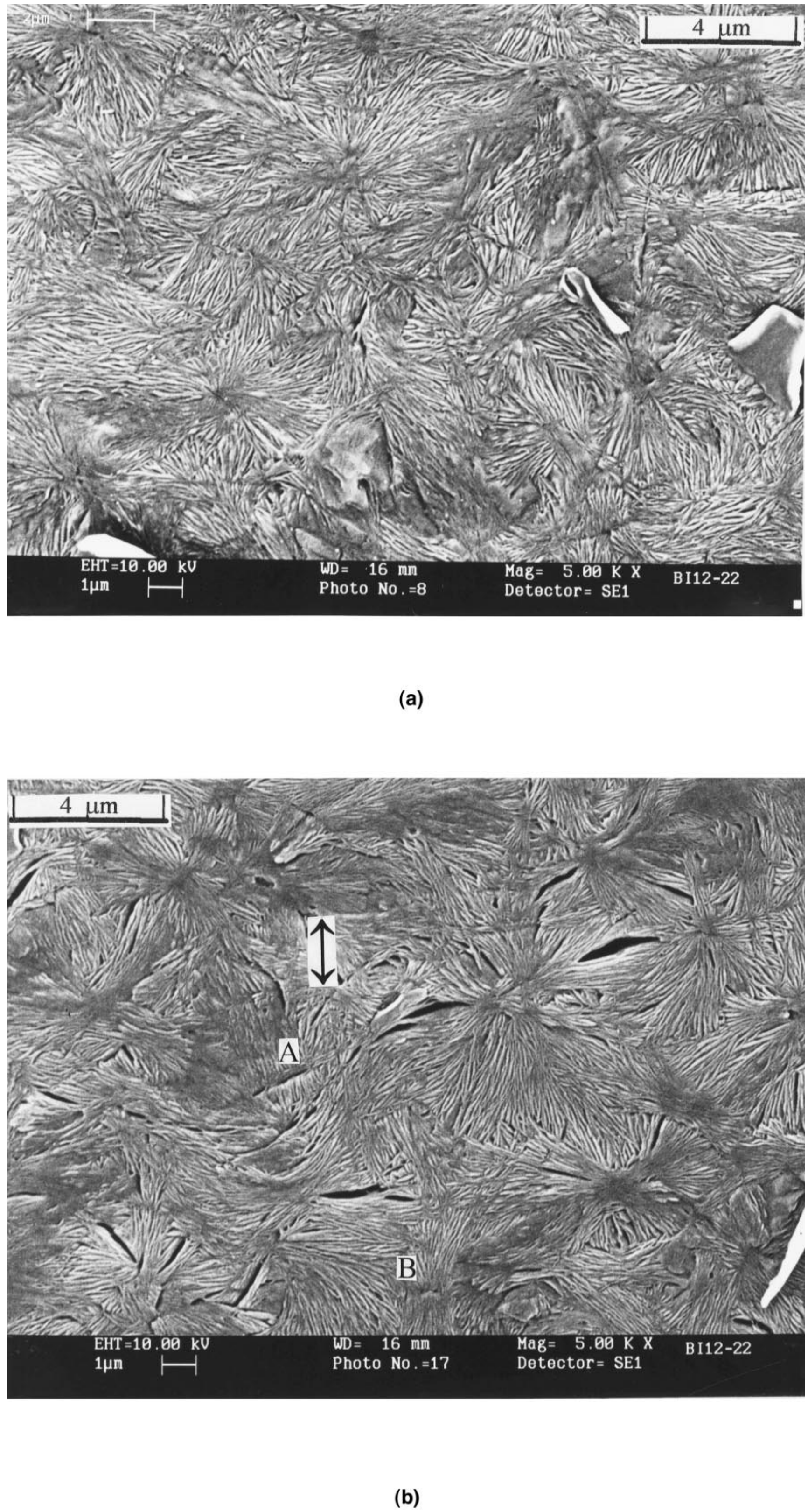


Fig. 1. SEM micrographs of injection molded β-PP sample: (a) undeformed sample; (b) sample at 5% strain inflicted by 90% of yield load.

From the view of microstructure, the bulk-crystallized plastics are complicated. On the one hand, they are composed of non-crystalline and crystalline regions (amorphous and lamellae). The response of the amorphous and lamellae to applied stress would be different. On the other hand, the lamellae may take any possible orientation within spherulites and the orientation of lamellae may affect their deformation behaviors. Even within the same lamella the deformation cannot be uniform because of the existence of defects in the lamella. Therefore, the deformation mechanism are certainly complex and multiple in the bulk-crystallized plastics.

In a previous work [11], a β-PP sample was examined with scanning electron microscopy (SEM) and transmission electron microscopy (TEM) before and after yielding. That sample possessed a high degree of crystallinity and exhibited uniform elongation within the whole work portion in macroscopic when it was stretched at room temperature. But the SEM and TEM examination revealed that the deformation of the specimen was highly inhomogeneous. Three primary deformation modes: (1) separation of lamellae; (2) inter-lamella shear and (3) intra-lamella slip, were identified in the deformed specimen. Separation of lamellae was a predominant mode in the early stage of deformation and most likely occurred in the area where the lamellae were perpendicular to the loading direction. Continuous separation of lamellae led to the formation of crazes and cracks within the spherulite upon further deformation. Near the yield point, intra-lamella slip was activated greatly in some areas where the lamellae were along the loading direction, resulting in local distortion and disintegration of lamellae. But the main cause of failure was cracking across the specimen.

In this present work, a β-PP sample was selected and stretched at room temperature. This sample has a lower degree of crystallinity than that examined before and could exhibit necking and cold drawing after yielding. The DSC and WAXS results [10] have shown that the β phase would transform to the α phase over the necking region. So the changes of lamella structure in the necking region could be examined with SEM and TEM. Hopefully, it will throw some light on the mechanisms of necking and cold drawing in bulk crystallized plastics.

2. Experiment

The β-PP specimens investigated were prepared by injection molding as reported before [10,12], which possessed an overall crystallinity of 58% and a percentage of β-phase about 61%. The specimens were tensile tested on a Lloyd LR 50 K tensile testing machine with a cross-head speed of 5 mm/min at room temperature. When the strain exceeded 8% (yield point) the specimens exhibited obvious necking and cold drawing phenomenon. After cold drawing several marks were made over the neck and the dimension of the cross-sections were measured under load. The actual strains of the materials at the labeled portions were calculated based on their draw ratio. After relaxation at room temperature for 6 months the specimens before and after yield were prepared for microscopy examination.

For SEM examination the specimens were first trimmed on the flank along the loading direction with a Leitz 1400 microtome and then were further trimmed with a glass knife on a Reichert-Jung Ultracut E microtome. The trimmed specimens were etched with permanganic acid in an ultrasonic bath [11]. After etching the specimens were coated with gold/platinum using a Bio-Rad coating system operating at 1.2 kV and 15 mA. SEM examination was conducted on a Cambridge S360 scanning electron microscope and the observation was made in the central position of the deformed specimens. During examination the stretched direction of the specimen was always arranged vertically in the view field. In this article the term of ‘vertical lamellae’ meant the lamellae parallel to the loading direction while the ‘horizontal lamellae’ represented the lamellae perpendicular to the leading direction.

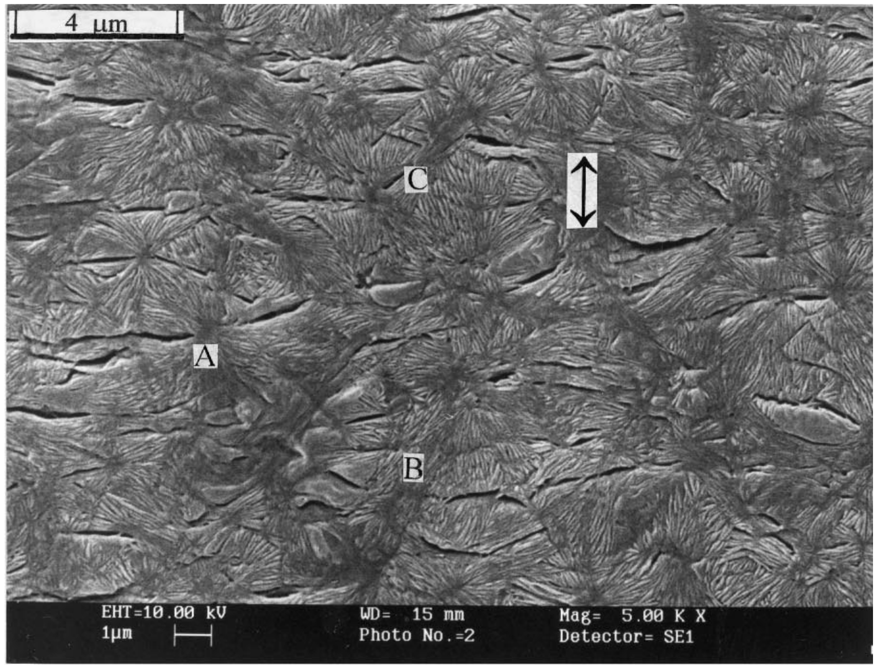
For TEM examination the deformed specimens were cut into thin strips perpendicularly to the loading direction. The cross-section of the strips was $0.2 \times 0.3\ mm^2$ and parallel with the flank of the tensile specimens. The strips were embedded in epoxy resin and cured at 40°C for 48 h. The embedded specimens were first trimmed on a Reichert-Jung Ultracut E microtome, and then stained in the vapor of ruthenium tetroxide [11,13] and cut into 50 nm thick sections. The ultrathin sections were mounted on 200 mesh copper grids and examined under a JEOL JEM-100SX transmission electron microscope operating at an accelerating voltage of 80 kV.

3. Results and discussion

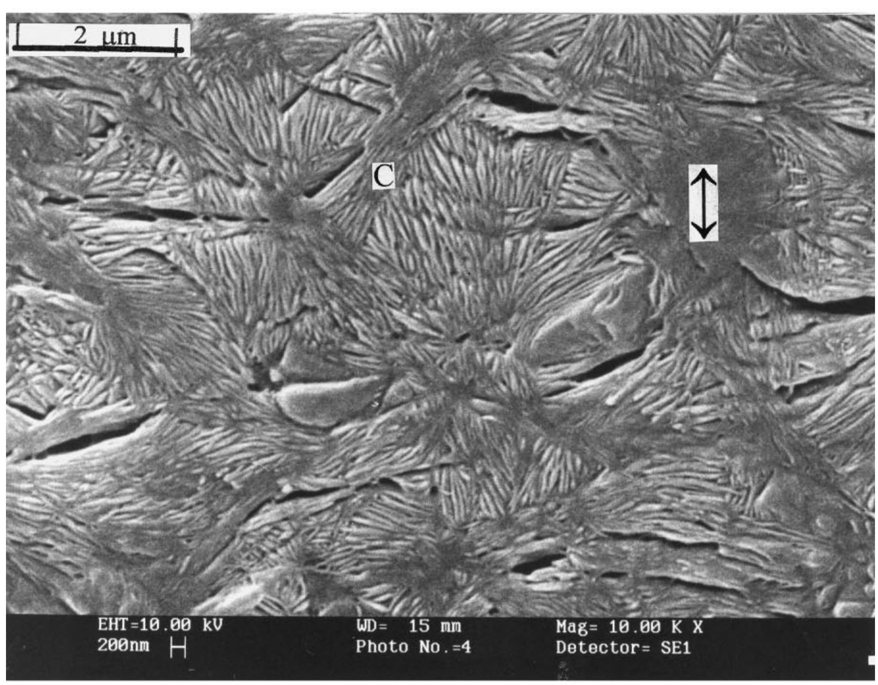
3.1. Morphology before yielding

Based on the growth fashion, the β-PP spherulite is classified in category II [14]. Normally, two symmetrical ‘‘eyes’’, the feature of category II spherulite, can be observed in the center of β-PP spherulite [15]. However, the mature category II spherulite could not be identified easily in this undeformed sample. Only a lot of sheaf-like lamellae, the immature spherulites, have been observed, as shown in Fig. 1(a). The general size of spherulites along their long axis is around 8 μm. Compared with the sample investigated before [11,15], which was prepared by compression molding at 130°C, the spherulites in this sample are much smaller. In fact, the spherulite in this sample could not develop well because the nucleating density was rather high due to the lower crystallization temperature and better dispersion of the nucleating agent in the injection molding process.

Fig. 1(b) shows a typical SEM view of the β-PP sample at

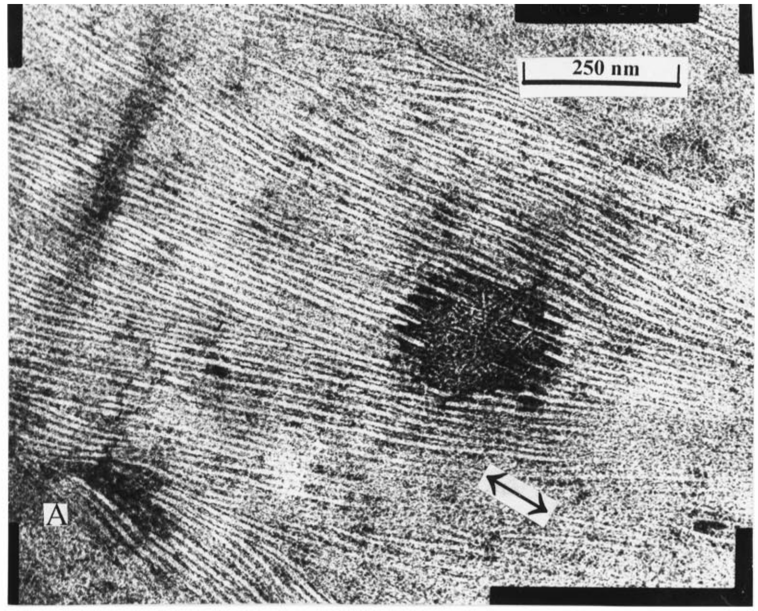


(a)

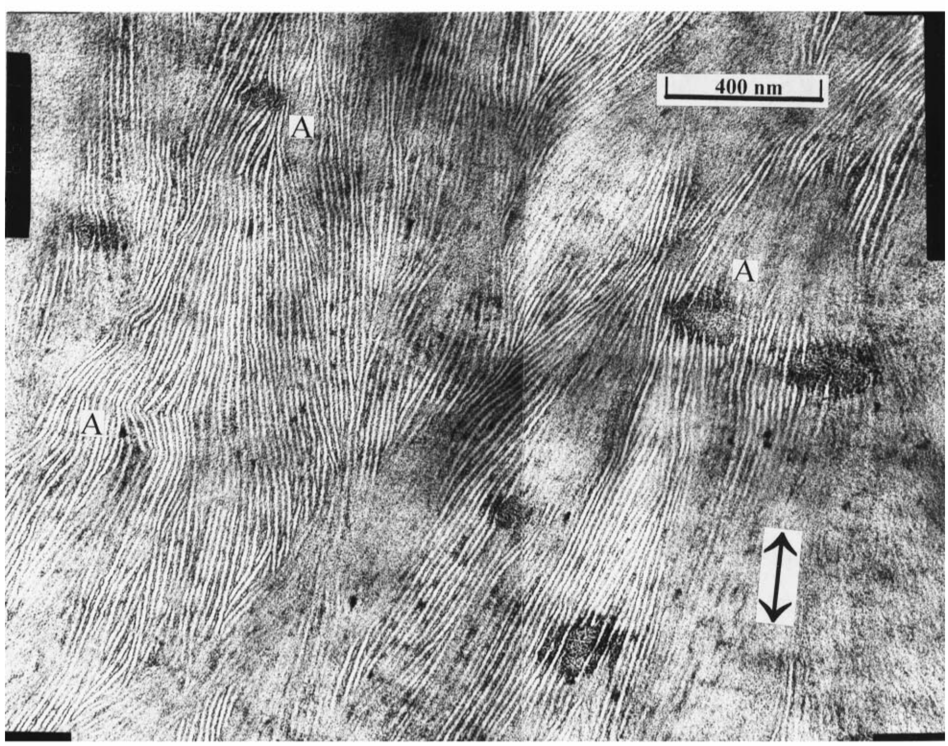


(b)

Fig. 2. SEM micrographs of injection molded β-PP sample at yield: (a) low magnification view showing crazes and deformation bands; (b) high magnification view showing shear of lamellae in the deformation bands.



(a)



(b)

Fig. 3. TEM micrograph of injection molded β-PP sample at yield: (a) melt spots with deformation band nearby; (b) area with dense melt spots.

5% strain which inflicted by 90% of its yield load. On the micrograph, the loading direction is vertical as indicated by the arrow. A few deformation bands or crazes (slits in Fig. 1(b)) could be found on the etched surface. Most of them were running along lamellae, nevertheless, there were a small number of slits across lamellae, as marked with the letters A and B. It should be pointed out that the original appearance of the deformation bands and crazes may look

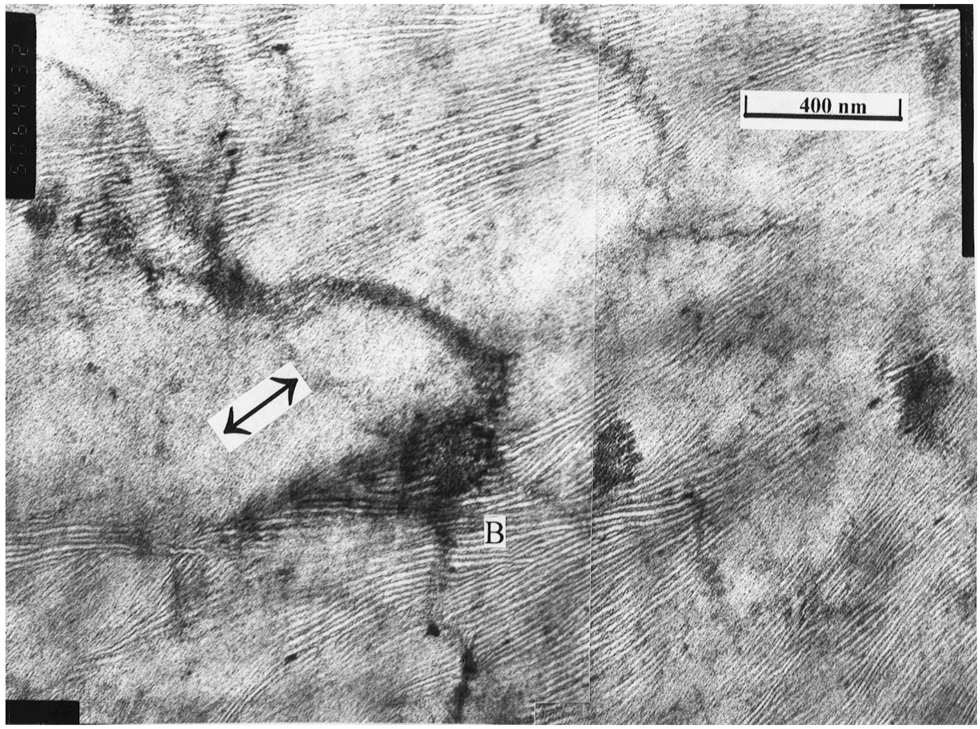


Fig. 4. Melt spots and deformation bands observed in yielded β-PP at 8% strain under TEM, showing the developing of melt.

different from the slits because some deformed materials were removed from them during etching. The observed result here is similar to the previous results on a β-PP with a higher degree of crystallinity [11]. However, the number of the deformation bands or crazes in this sample is larger but the size is smaller at the same percentage of yield load. Some slits were 3 μm long and running almost half way across the spherulite from the center to the periphery. Additionally, there were more crazes or deformation bands inclined to the loading direction. In glassy polymers, crazing normally occurs in a perpendicular direction to the applied tensile load. The formation of crazes inclined to the loading direction in β-PP is probably due to the combined effect of the lamella orientation and local stress. Although the applied tensile load was unidirectional (vertical), it is possible that local stress was generated in other directions as a result of inhomogeneous deformation within the material. The local stress may cause inter lamellae shear and separation of lamellae in the areas where the lamellae are inclined to the loading direction.

3.2. Morphology of yielded sample at strain 8%

As the strain was increased to 8%, the specimens began to yield; further straining would have led to necking and cold drawing. Fig. 2 shows a SEM view of a yielded specimen but without obvious necking. More crazes along lamellae appeared in the specimen and some crazes have coalesced together across neighboring spherulites, as seen in area A of Fig. 2(a). Apart from crazing along lamellae, some fine deformation bands across vertical lamellae (dark lines marked with B and C) were also observed. They took a similar orientation as the crazes roughly perpendicular to the loading direction but appeared as slight depressions on the etched surface instead of open. The deformation bands (dark lines) reflected a local distortion of the vertical lamellae. At higher magnification (Fig. 2(b)), it can be seen that the lamellae in the deformation bands became thinner and appeared to have undergone local ‘‘necking’’. Furthermore, there was a slight shear displacement between the upper and lower parts of the lamellae. Such shear deformation likely involved intra-lamella slipping along the ‘‘*c*’’ axis of polymeric crystals, as proposed by Bowden and Young [2].

Previous work [11] on a β-PP with a higher degree of crystallinity has shown that in the yielded specimen some melt spots could be found among the lamellae parallel with the loading direction. In this present work melt spots have also been observed in the yielded specimen under TEM. Fig. 3 shows TEM micrographs of the yielded specimen at 8% strain. The arrow on the micrograph indicates the loading direction. A number of melt spots can be seen among lamellae. Major melt spots appeared near the branching sites of lamellae as marked with A. Furthermore, some melt spots with deformation bands near-by have been observed in the yielded specimen (Fig. 3(a)). Such deformation bands closed to melt spots were scarcely found in the previous work [11]. In comparison with the previous work, the melt spots are smaller but more plentiful at a similar stress level. This indicates that the lamellae in this sample can be destroyed more easily. Perhaps because the lamellae in this

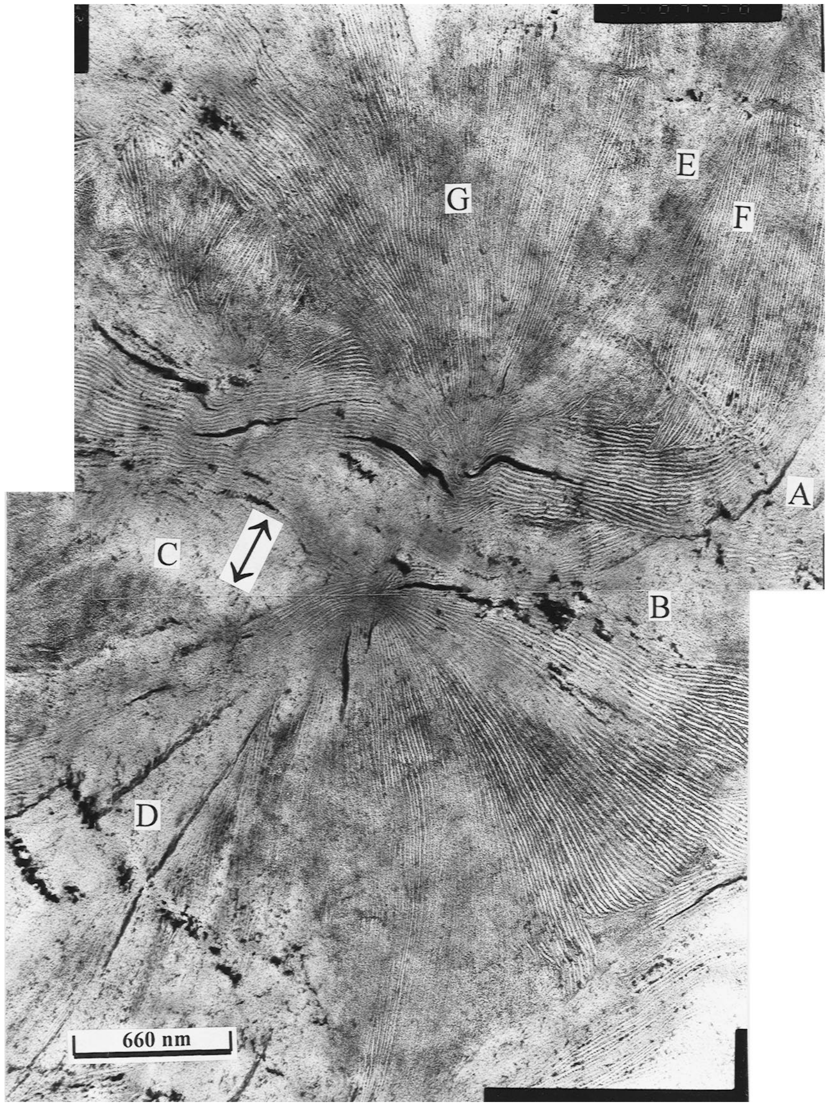


Fig. 5. β-PP spherulite observed in yielded specimen at 8% strain under TEM. A, B, and C: heavily distorted regions; D: crazes near pole of spherulite; E: deformation bands near pole of spherulite; F, G, and H: slightly distorted lamellae.

sample are less perfect than those in the previous work and are easy to distort, therefore, the number of possible sites for melting is larger. As a result, less energy is involved in each site, hence, the melt spots are smaller. Fig. 4 is another TEM micrograph showing melt spots and deformation bands. From the thickness variation of the deformation band through the center, it has probably been initiated from one of the melt spots (B). Furthermore, the melting process seems to have spread from the melt spot along the lamellae towards the left.

Fig. 5 is a TEM micrograph of the yielded specimen which shows various deformations within a β-spherulite. If the applied stress is considered to act along a line joining the two poles of the spherulite, the lamellae in the equator region of the spherulite are perpendicular to the loading direction while those in the pole regions are parallel with the loading direction. It can be seen that the phenomenon of crazing along lamellae is rather apparent, especially in the equator region. Also, a number of areas (A, B and C) appear to have been distorted heavily and the lamella structure can no longer be recognized. In the poles of the spherulite, there are some continuous deformation bands across lamellae. They have an orientation perpendicular to the loading direction. One may have noticed that the gray of these in the low pole (bottom left corner of Fig. 5) is deeper than those in the upper one. This reflects that those in the low pole were deformed more due to higher local strain. Perhaps, they are crazes developed from deformation bands similar to the upper ones. In the vicinity of the crazes in the low pole, there are a few dark bands running parallel with the lamellae. They might be initiated by inter-lamellae shear due to local stress generated more or less along the lamellae. At a later stage, such deformation will inevitably involve the crystals and it can be seen that some lamellae next to band D have been affected. There are no apparent melt spots in this micrograph, nevertheless, a few dark marks can be seen near

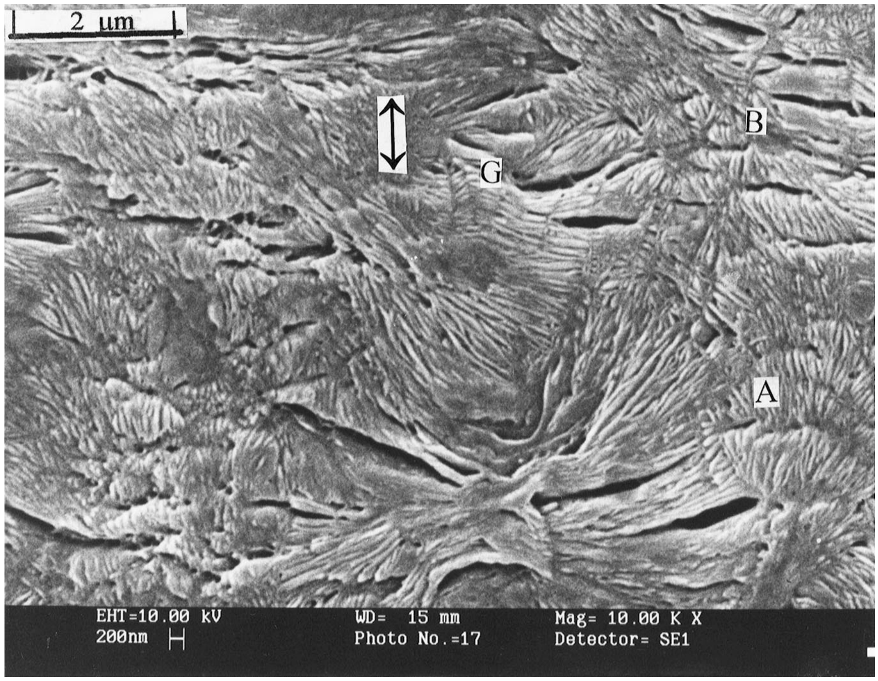


Fig. 6. SEM micrograph of β-PP at the early stage of necking. A: coarse deformation bands across vertical lamellae; B: crazes across vertical lamellae; G: shear bands along the loading direction in horizontal lamellae.

the area around E. Perhaps, these dark marks are signs of melt spots which were sectioned near the periphery of melt spots, thus the appearance is not so clear.

In the area just to the left of the heavily deformed region A, the original orientation of the lamellae should be horizontal but now the section next to the heavily deformed material has been bent upwards to align parallel with the applied tensile load. Furthermore, the bent sections of the lamellae became thinner than the unaffected sections. Perhaps, the bending and thinning phenomenon resulted from inhomogeneous deformation in the material. The material at region A has suffered a substantial local strain along the loading direction whilst the strain is relatively small in the horizontal lamellae to the left of region A. As

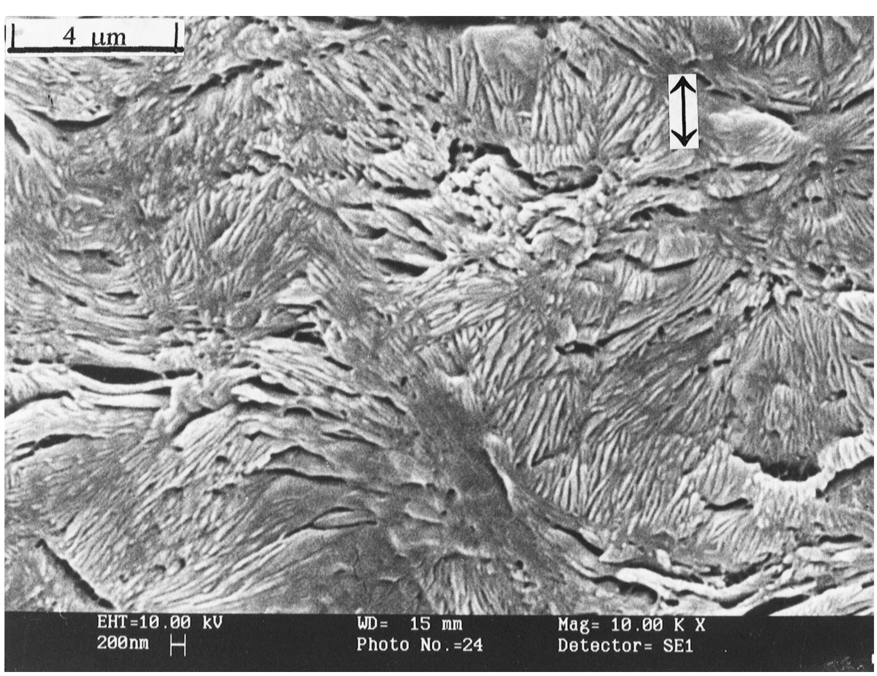


Fig. 7. SEM micrograph of β-PP at upper shoulder of neck, showing extensive distortion of lamellae.

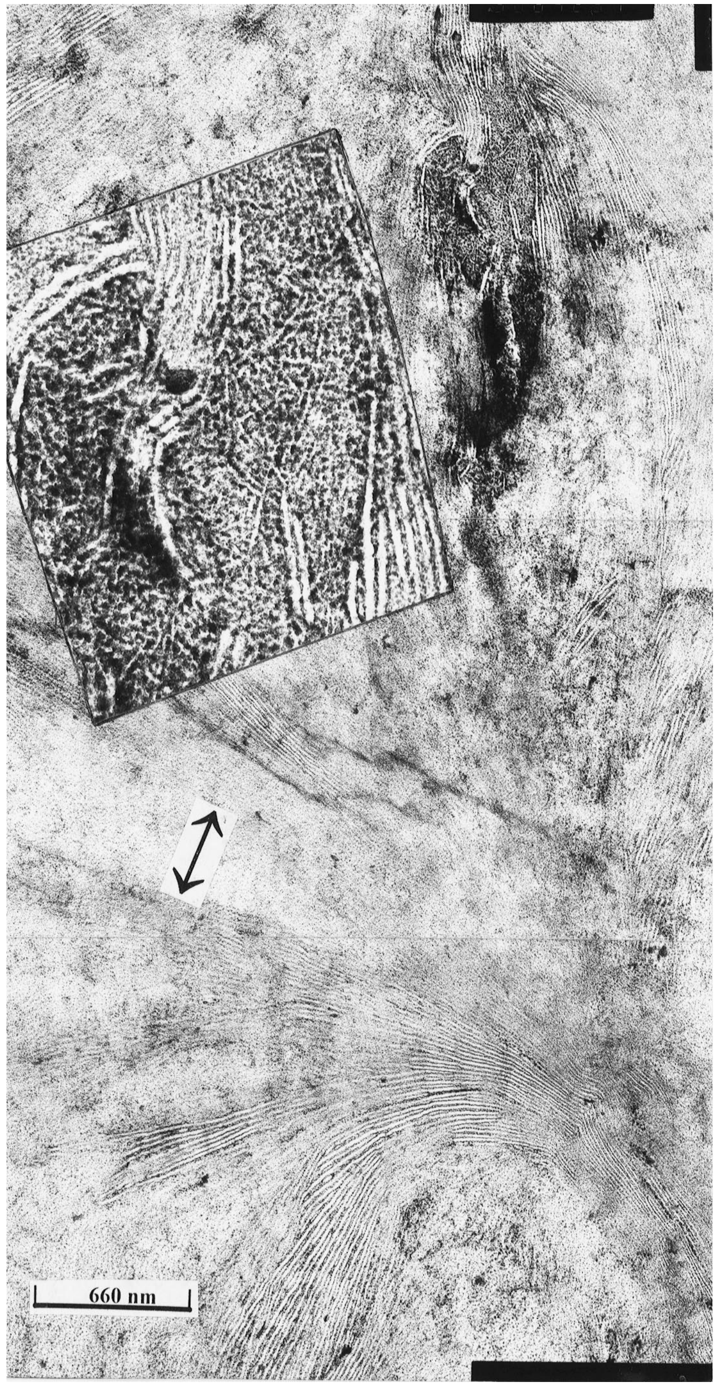
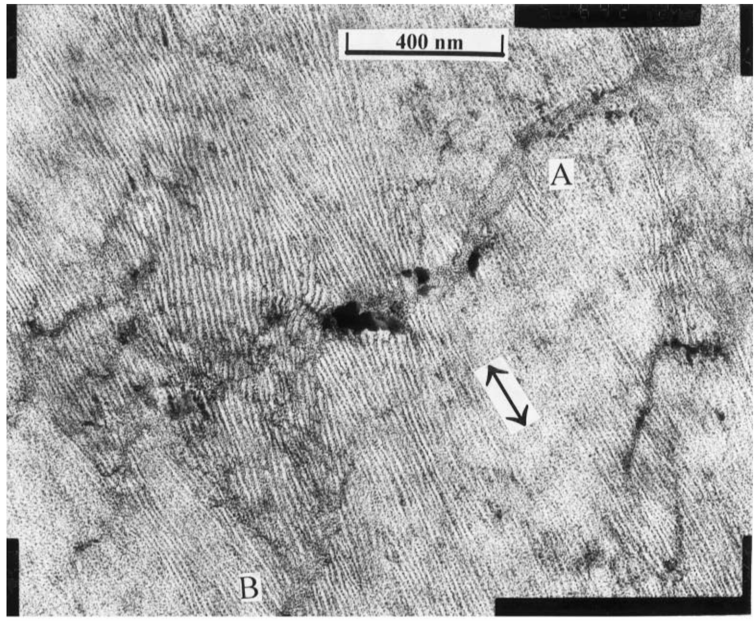


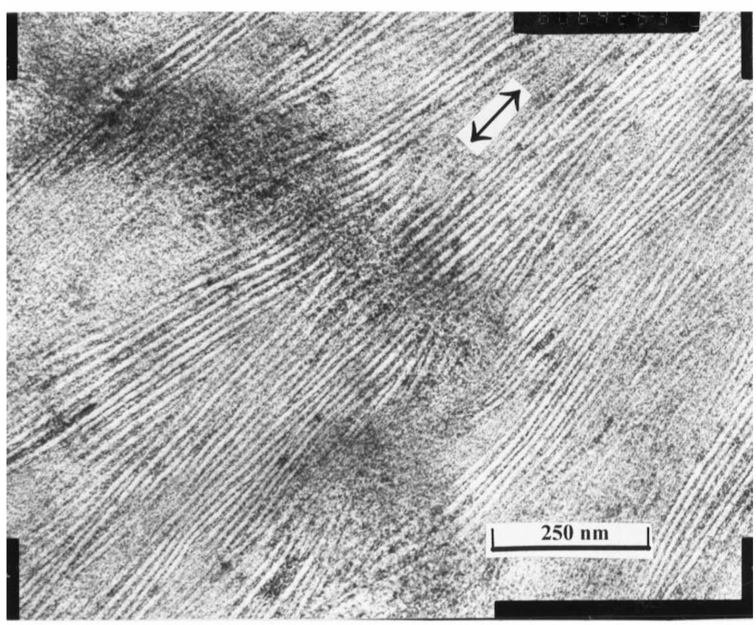
Fig. 8. Elongated melt spot observed in the upper shoulder of neck of β-PP at 22% strain under TEM.

a result, a shear lag is present between the two regions, thus a shear stress is generated along the loading direction and it will cause the lamellae to rotate. The neighboring lamellae have to adjust their relative position to accommodate their different radii of curvature at the bend and this can be achieved by inter-lamella shear. On the other hand, the reduction of the thickness of the tilted lamellae sections can be attributed to intra-lamella slipping. In addition, the lamellae in regions F and G are also thinner than usual. It is likely that they have suffered a substantial longitudinal strain through extensive intra-lamella shear and slip tilting process.

In comparison with the previous work [11], the yielded sample contained more melt spots and deformation bands across the vertical lamellae. Furthermore, extensive intra-lamella slip and obvious rotation of lamellae were found in this yield specimen. This observation seems to indicate that the lamellae in the present sample could be distorted and destroyed more easily than those in the previous work. Obviously, this was attributed to the defects of the crystals



(a)



(b)

Fig. 9. TEM micrograph of the upper shoulder of neck of β-PP at 22% strain under TEM: (a) networks of deformation bands; (b) coarse deformation bands.

in the present sample. Actually, DSC results [10,12] have shown that the sample used in this work has a lower crystallinity, wider melting range and lower melting point. Hence, intra-lamella slip and ‘‘melting’’ under the action of the tensile load are relatively easier, particularly in the less perfect parts of lamellae. It should be pointed that the phenomena of extensive thinning of vertical lamellae and rotation of horizontal lamellae are not obvious in the previous work [11].

3.3. Morphology at an early stage of necking

Fig. 6 shows the morphology at the beginning of neck with a local strain of about 20%. At the early stage of necking, deformation bands and crazes became more apparent in all areas regardless of lamella orientation. Compared with the sample at yield, the vertical lamellae were deformed more seriously. In the areas of vertical lamellae, besides the fine deformation bands, some coarse deformation

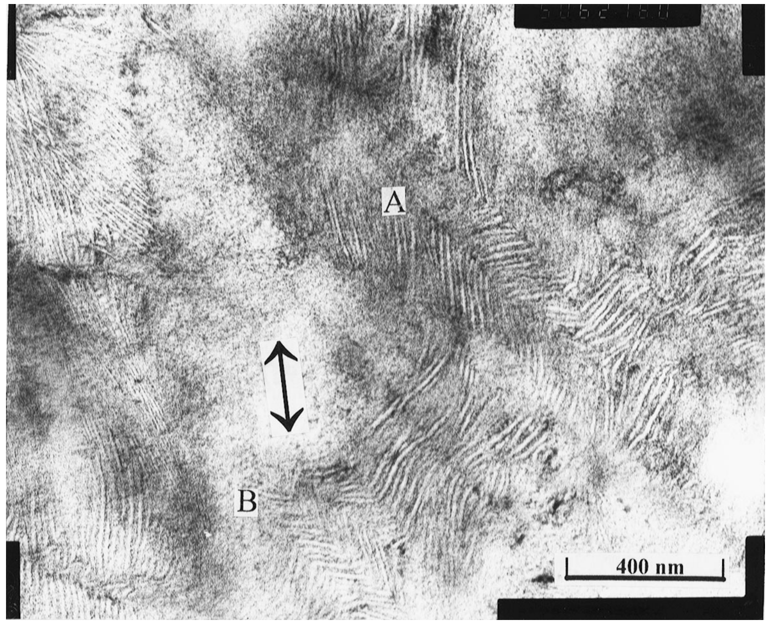


Fig. 10. TEM micrograph observed in the tapered section of the neck of β-PP at 35% strain, showing shear bands along the loading direction.

bands and crazes across the vertical lamellae could be observed (as marked with A and B of Fig. 6). Furthermore, many of the lamellae were broken into short fragments (on the left of Fig. 6). In contrast, the horizontal lamellae seemed to be less affected. This is probably due to the fact that they are parallel with the crazes and most of the deformation was taken up by crazing within the amorphous materials. Somehow, vertical shear bands could be observed occasionally in the horizontal lamellae, as marked with G in Fig. 6, the horizontal lamella bundle were broken and tilted towards the loading direction. This was believed to be due to the higher strain in the adjacent vertical lamellae along the loading direction.

3.4. Morphology at upper shoulder of neck

Fig. 7 shows the morphology observed at the upper shoulder of a fully developed neck, i.e. the zone just below the parallel section of the gauge length and the corresponding strain is 22%. The morphology of the upper

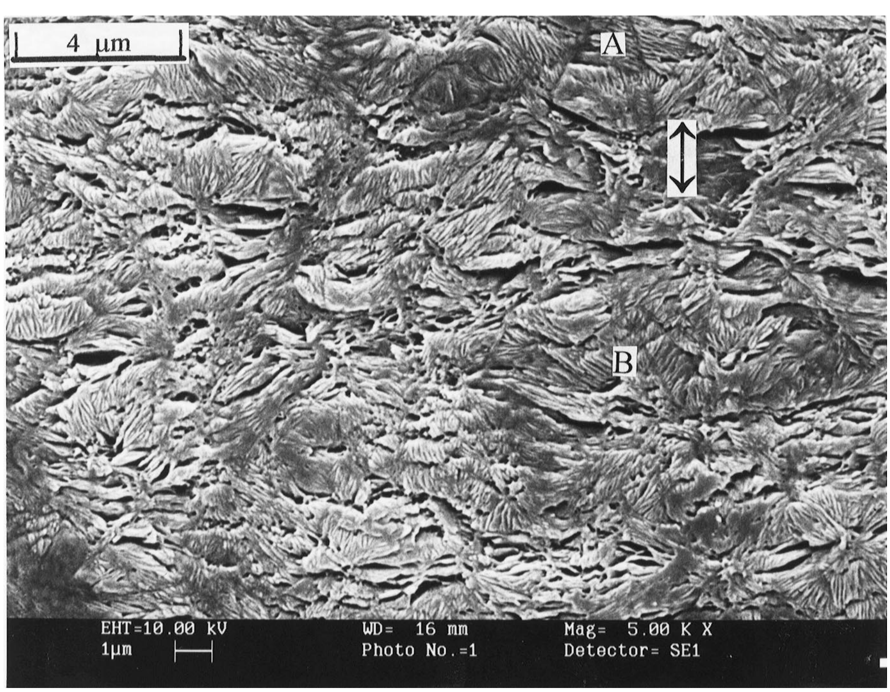


Fig. 11. SEM micrograph of β-PP at the tapered section with 50% strain. The spherulites were shattered.

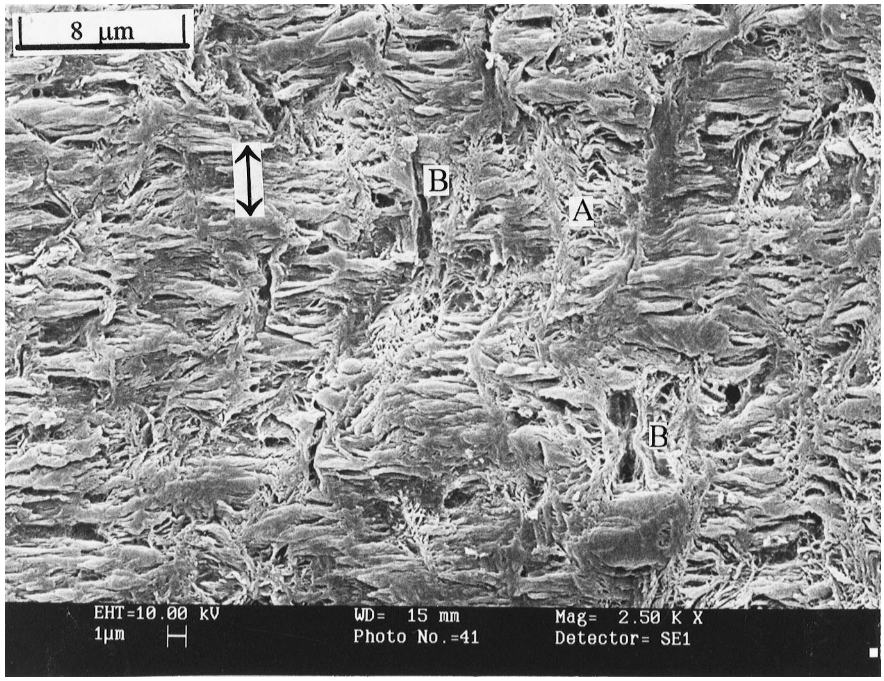


Fig. 12. SEM micrograph of β-PP at the tapered section with 80% strain: (A) material flow in the loading direction; (B) longitudinal pits.

shoulder is similar to Fig. 6 of the early stage of necking but the lamella structure became less sharp, reflecting the high degree of deformation within the material. One may see the area near the top left corner of Fig. 7, although segments of lamellae can be identified, however, the SEM is no longer able to resolve the fine details of the lamellar structure. It was believed that the lamellae within such an area has been seriously distorted.

The upper shoulder of neck was also examined under TEM. In the upper shoulder melt spots, as shown in Fig. 3, were observed more often. Moreover, large elongated melt spots could be found in this section. Fig. 8 shows such a melt spot observed in the upper shoulder of the neck at strain 22%. There are two lamella sheaves from two spherulites on the top and bottom of the micrograph. The elongated melt spot was located near the poles of the adjacent spherulites. Under close examination, a number of fine white strips could be seen interwoven together within the melt spot. They are believed to be α-phase crystals which have recrystallized from the melt. In fact, previous

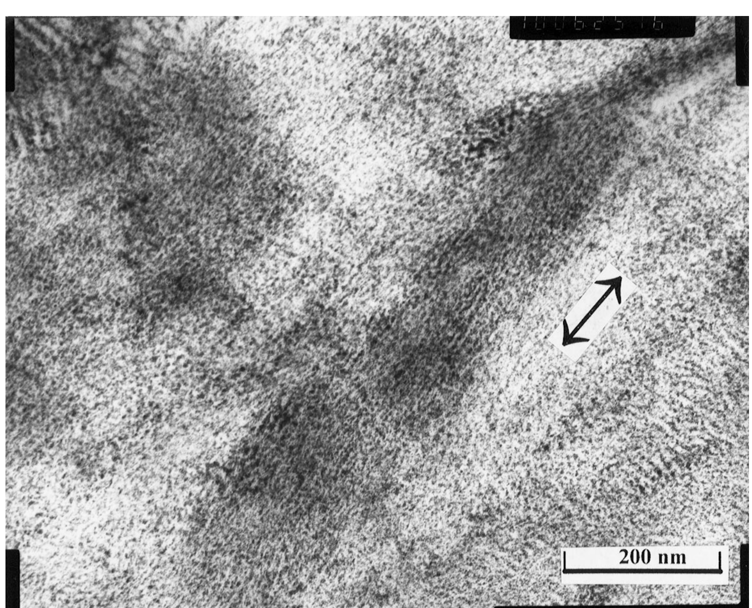


Fig. 13. TEM micrograph of β-PP at the tapered section with 80% strain, showing microfibrils and fragment of β-lamellae.

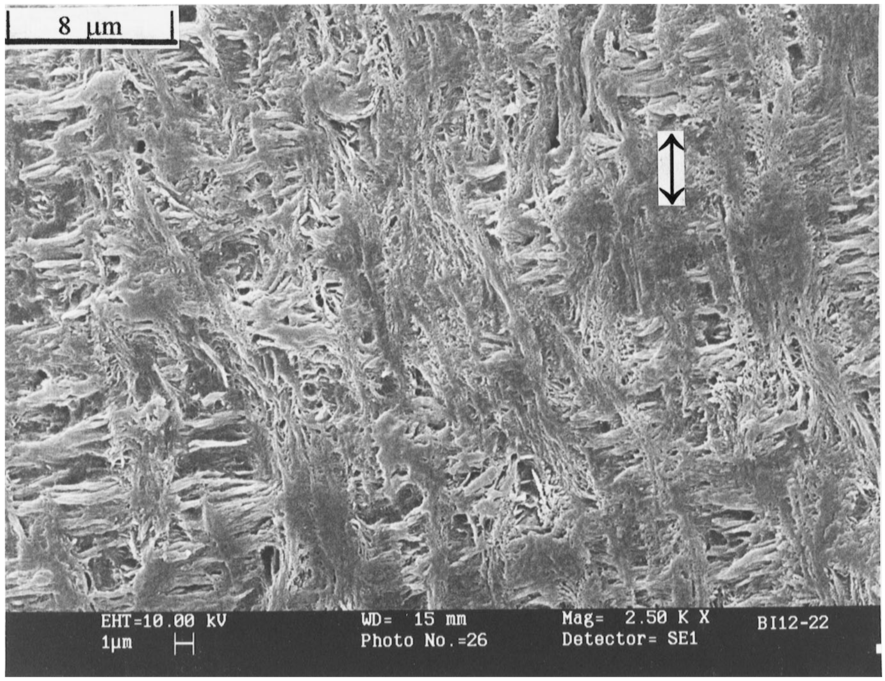


Fig. 14. SEM view of β-PP after cold drawing to 170% strain.

DSC examination [10] indicated that in the upper shoulder of the neck at 22% strain, the crystallinity of the β-phase dropped from 36 to 33% whilst the α-phase increased from 23 to 26%. Most of the fine long crystalline strips within the elongated melt spot were aligned parallel with the loading direction and they are believed to be the strain-induced crystals. Furthermore, the area below the melt spot appears darker due to absorption of more staining agent. This is a sign of heavy distortion of the material. Perhaps, it will melt and recrystallize on further straining and become part of the enlarged melt spot.

Besides the melt spots, a lot of deformation bands were observed in the upper shoulder of the neck under TEM. Some deformation bands were rather fine but were crowed in a local region. They were connected to each other and looked like a web, as shown in Fig. 9(a). Coarse deformation bands were also observed in this section; some of them were about 200 nm wide and more than 1 μm long. They

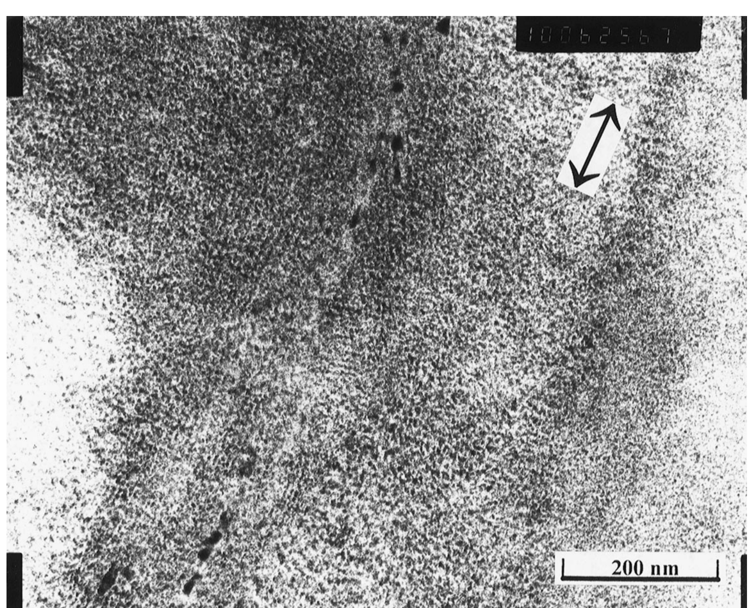


Fig. 15. TEM Micrograph of cold drawn material of β-PP at 160% strain.

took an orientation perpendicular to the loading direction and across lamellae, as shown in Fig. 9(b). In some local sites within the coarse deformation band the gray of the bands was similar to that of the melt spot. Perhaps, melting had occurred within these bands locally. It should be pointed out that such deformation networks and coarse deformation bands were not found in the previous work [11] on the sample with a higher degree of crystallinity. This was probably due to the fact that the crystals in the previous sample were stronger and not easily deformed.

3.5. Morphology at the tapered section of a fully developed neck

The cross-section of the neck dropped rapidly below the upper shoulder of the neck and the calculated strain changed from 30 to 80% within a length of less than 2 mm. Fig. 10 shows a TEM micrograph within the tapered section where the strain is about 35%. Two shear bands are running almost parallel with the loading direction. It can also be seen that the broken lamella segments within the shear bands have tilted closer to the loading direction. These shear bands are in fact similar to those observed at the early stage of necking (ref. G in Fig. 6). In general, almost all the lamellae have suffered a certain degree of deformation, the vertical lamellae being stretched thinner while the horizontal ones being sheared and tilted towards the loading direction. Fig. 11 shows the morphology of another section where the strain was about 50%. The spherulites were shattered, no spherulite outline could be identified on the specimen. Nevertheless, some broken lamellae could still be seen. Furthermore, there are a number of fine deformation bands included in the loading direction (see A and B). These bands are believed to be of the same nature as the shear bands in Fig. 10.

As the strain was increased to 80% in the neck, the sample lost the original structure feature completely and the lamella-spherulite structure was no longer visible, as shown in Fig. 12. The specimen contained a large number of lamella fragments. These fragments were perpendicular to the loading direction and separated by material flows (marked with A). In addition, some longitudinal pits (marked with B) were observed in this section. These pits are actually cracks because such a kind of pits could be observed even on the trimmed specimen before etching. Perhaps, as necking proceeded, many shear bands along the loading direction were formed in the horizontal lamellae, thus more horizontal lamellae were broken up into fragments. Also, more vertical lamellae became thinner due to intra-lamella slipping and possible ‘‘melting’’. This gave rise to a plastic flow of material generally along the loading direction. Meanwhile, some of the horizontal lamella bundles rotated towards the loading direction as the adjacent material ‘‘flowed out’’, which thus gave a more favorable orientation for intra-lamella slipping and fed into the plastic flow. On the other hand, accompanied with the rotation of lamellae, some crazes formed between lamellae in the early stage of deformation might change their orientation with the lamellae together, as a result, they might become more inclined to the loading direction.

Fig. 13 shows a TEM micrograph of the section with a local strain of 80%. Apart from a few β-lamella fragments at the top left corner, the original lamella structure has largely been destroyed. The region is probably within a vertical material flow such as A in Fig. 12. Traces of very fine crystals can be seen along the loading direction. Since the DSC and WAXS results [10] have shown that most of the β-phase would transform into α-phase at this section, they are likely to be α-phase recrystallized under the applied stress.

3.6. Morphology of cold drawn material

Fig. 14 shows the morphology of a cold drawn section at 170% strain. The volume of horizontal lamellae fragments has decreased in the cold drawn material. In fact, most of them were consumed by the plastic flow in the vertical direction on drawing. Fig. 15 shows the morphology observed on another cold drawn section under TEM. Despite the tensile strain of this section (160%) being much higher than that of the tapered section in Fig. 13, there is little difference between their morphologies. The phenomenon can be attributed to the discrepancy between the calculated strain and the actual strain in the tapered section. Although the overall tensile strain of the tapered section is about 80%, the actual strain within the vertical material flows is much higher, whilst the strain of the horizontal fragments is lower due to inhomogeneous deformation. The highly drawn section in Fig. 13 is probably located in a vertical material flow and its true strain is much higher than the overall calculated value of 80%. In contrast, most material in the cold drawn section has been deformed and the calculated strain more or less reflects the true strain across the whole section.

4. On deformation mechanisms

In a previous study [11] on a β-PP sample with a higher degree of crystallinity, it was observed that the strains were highly inhomogeneous, although the specimen deformed uniformly in the whole gauge length. In the early stage of deformation, the strain was mainly accommodated between horizontal lamellae, led to the formation of deformation bands or crazes along lamellae. Near the yield point, some vertical lamellae were stretched to broken, which resulted in continuous deformation bands or local disintegration of the lamellae. However, the main cause of failure of that sample was coalescence of crazes along horizontal lamellae and cracking across the specimen.

In this present work, it has been observed that the structure changes before yielding are similar to those observed before [11]. The strain was mostly due to deformation in the amorphous layers between the lamellae. Nevertheless, more deformation bands inclined to the loading direction were observed. Strictly speaking, not all molecular chains within these inter-lamella layers are arranged in random, but some partially ordered domains may exist. Extension of molecular chains in the inter-lamella layers would cause destruction of the partially ordered domain, especially in the region where the lamellae are perpendicular to the loading direction. Perhaps, fatigue-softening of crystalline plastics can be interpreted with the term of destruction of the ordered domains because the number of ordered domains (physical entanglements) in the inter lamella layers will reduce during circle loading even at a low stress level.

At the yield point, besides crazes along the horizontal lamellae, many melt spots and deformation bands across vertical lamellae appeared. Furthermore, some vertical lamellae became thinner through extensive intra-lamella slip and some horizontal lamellae rotated towards the loading direction. The phenomena of extensive thinning of vertical lamellae and rotation of horizontal lamellae were not obvious in the previous work [11] on a sample with a higher degree of crystallinity and they are believed to play an important role in the necking and cold drawing of crystalline plastics. Whether a semicrystalline plastic will neck and cold draw or break in a brittle fashion under a given condition depends greatly on the ability of the crystals to deform.

At the beginning of necking, the melt spots were enlarged and elongated in the stress direction, meanwhile strain-induced recrystallization occurred in the melt phase. Moreover, more vertical lamellae were affected while the horizontal lamellae remained less affected, more fine deformation bands across vertical lamellae appeared and some grew into coarse deformation bands, the vertical lamellae were broken into short fragments. Somehow, a few shear bands included the loading direction generated in the horizontal lamellae when the adjacent vertical lamellae were stretched heavily, thus, the horizontal lamella bundle was broken and tilted towards the loading direction.

As necking continued, the spherulites were broken up into fragments by deformation bands and crazes. At the deflection section of the neck, the spherulites were shattered completely. On the other hand, excessive intra-lamella slip led to the vertical lamellae to be thinner, disintegrate, flow and recrystallize, as a result, some highly drawn material domains were formed. In turn, shear bands were generated roughly along the loading direction and more horizontal lamellae were broken up into fragments; the broken sections rotated towards the loading direction and fed into the material flows continually. Finally, the lamella–spherulite structure was converted into an oriented fibril structure in the cold drawn material.

The deformation mechanisms of semicrystalline plastics are certainly diverse and complex, especially for bulk crystallized plastics. Based on the examination of the structure of β-PP over the necking region and combined with previous work [10,11], however, the deformation mechanisms of necking and cold drawing in the bulk crystallized β-PP can be depicted as follows. The yielding of PP is mainly concerned with the motion of the chain segments [10] and the primary deformation modes include [11]: (1) separation of lamellae, (2) inter-lamella shear, and (3) intra-lamella slip. Inter-lamella separation is likely to occur near the equatorial planes of spherulites where the lamellae are perpendicular to the loading direction. In contrast, intra-lamella slip takes place mostly in lamellae along the loading direction. Before necking, intra-lamella slip mainly operates near the poles of the spherulite, while after necking it also becomes operative in the rotated section of the horizontal lamellae.

When a uniaxial tensile load is applied on β-PP, in the first stage of deformation, the strain mainly involves the inter-lamella layers. The amorphous material between the horizontal lamellae are stretched, resulting in separation of lamellae and even crazing along the lamellae. At this stage, the stress–strain relation generally obeys Hooke’s law. In the secondary stage, as the strain approaches the yield point, separation of lamellae continues and more deformation bands and crazes generate near the equatorial planes of the spherulite. Meanwhile, inter-lamella shear becomes obvious in the regions where the lamella are inclined to the loading direction. Also, intra-lamellae slip may occur on the lamellae parallel with the loading direction. Some of the vertical lamellae are stretched to thinning, local necking or even disintegration, resulting in the formation of continuous deformation bands and melting spots. The breaking and local melting of vertical lamellae contribute to the yielding of material when stretched at room temperature. At this stage, the modulus of material descends rapidly and the stress–strain curve deviates from linear response.

In the third stage of deformation the predominant deformation mode may be different, depending on the structure of the material itself and deformation conditions, such as strain rate and testing temperature. If the melt-crystallized sample has a lower degree of crystallinity and the lamellae contain more defects, or the strain rate is slow enough to allow the movement of the chain segments under the tensile load, then intra-lamella slip will dominate. Dense deformation networks, coarse deformation bands and elongated melt spots will form, especially near the poles of spherulites. Furthermore, the inclined oriented lamellae become thinner and extend along the loading direction. Meanwhile, the nearby horizontal lamellae rotated towards the stretching direction through the combined effect of inter-lamella shear and intra-lamella slip. All these phenomena are believed to be contributing factors to initiate the necking process. As the strain is further increased, the horizontal lamellae will be sheared into fragments and the broken fragments rotate towards the loading direction. The rotation renders the lamella sections a more favorable orientation to disintegrate and add to the material flow as the neck propagates.

However, if the melt-crystallized sample possesses a high degree of crystallinity and lamellae are more perfect, or the strain rate is too high to allow the movement of the chain segments, separation of lamellae becomes the predominant mode of deformation. In this case, crazing occurs along the horizontal lamellae through extension and finally scission of molecules in the inter-lamella layers due to high local stress. On the other hand, slender deformation bands across the vertical lamellae cannot grow much wider or develop into deformation network but classical crazes. Finally, crazes develop perpendicularly to the loading direction and the material fails through cracking across the specimen without necking and cold drawing.

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