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Propagation of shear bands through obstacles in atactic polystyrene

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High speed cinematography (up to 6000 frames sec^{-1}), as well as optical and electron microscopy, was used to study the formation and propagation of thin and thick shear bands in atactic polystyrene, with and without obstacles in their paths of propagation. Three different kinds of obstacles were studied, an existing thick band, a fully recovered thick band and a region dispersed with 2 to 6 μm rubber particles (a strip of high impact polystyrene). Except for the fully recovered thick band which behaved like undeformed polystyrene, the other two obstacles effectively reduced the speed of propagation of the shear band packets and changed their mode of operation by dispersing them into thin bands and spreading them out into larger spaces. However, the original localized mode of operation resumed after the shear band packet passed through the obstacles.

1. Introduction

When atactic polystyrene (PS) is compressed, air cooled specimens develop coarse shear bands at stress concentrations under high strain rates and quenched specimens develop fine bands (or shear zones as observed in the light microscope) under low strain rates [1, 2]. While the coarse bands propagated at high speed in a localized region, fine bands spread by repeated nucleation around the zone. When the strain rate is very high (~ 0.1 sec^{-1}) thick bands appear [3] as a result of combination of numerous (thin) coarse bands in a band pocket. The thick bands are basically similar to coarse bands except that they are 1000 times thicker and have 50% more shear strain than the (thin) coarse bands. Because of their thickness, the thick bands reveal their fibrous structure more clearly than the thin bands which show striations only at large magnifications. Microcracks between fibrous strands frequently occur in thick bands but never in thin bands.

Thin coarse bands were observed to propagate through other coarse bands with difficulty. They shear each other in all three types of intersections as reported [4]. Microcracks and voids were produced at the intersections of band packets [5]. The propagation of one band packet was hindered by the other band packet. In order to understand further the propagation behaviour of shear bands through obstacles, existing thick bands, fully recovered thick bands, and regions of dispersed rubber particles were introduced as obstacles in the path of the propagating bands. High speed cinematography at up to 6000 frames sec^{-1} was used for the recordings and measurements of the propagation velocity of coarse bands and band packets.

Shear band growth rates at constant load were studied by Kramer [6] using a time-lapse camera. The velocities reported were in the range of 10^{-3} to 10^{-5} cm sec^{-1}. According to his description and the findings of Wu and Li [1, 2], the velocities he reported were for fine bands which grew by spreading. The velocities reported in this work are in the range of 0.2 to 20 cm sec^{-1} and are for coarse bands which propagate at high compressive strain rates. Fine bands and coarse bands represent different slip processes, as reported before [1, 2]. The present measurements and observations will reveal further the nature of localized deformation in polymers.

2. Experimental procedure

2.1 Sample preparation

Sheets of atactic polystyrene, about 6 mm thick, were obtained from the Westlake Co. It was the

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same material as has been used previously [1–5]. Blocks cut from the sheets were annealed for 20 h at 115° C and furnace-cooled slowly to room temperature over a period of about 6 h. Specimens were cut from the blocks in a milling machine. They were polished to 0.05 μm alumina finish and were annealed again at 98° C for 16 h and then again furnace-cooled slowly to room temperature.

2.2. High speed cinematography

Specimens were compressed in an Instron testing machine at a high strain rate of about 0.12 sec^{-1}. The compressive displacements were controlled by using cross-head control dials. Since shear bands usually propagate at high speeds, a Radlake Hycam model 41 16 mm high speed movie camera was used for the study of the shear band propagation. The camera speed used ranged from 500 to 6000 frames sec^{-1}. For larger magnifications, spacers of different sizes were inserted between the lens and the film. A 300W small tungsten bulb with a Dichroic reflector which absorbed infra-red radiation was placed at about 25 cm away from the specimen as the only source of illumination. The intensity of the light was increased and/or a larger aperture was used as the exposure time was decreased. For a camera speed of 500 frames sec^{-1} with an $f11$ camera aperature, the voltage for the light bulb was set at 80 V on a 5 amp Variac. The light was turned on only a few seconds before the test so as to minimize the heating effect. An air blower was also used during the tests. The test usually lasted only a few seconds. The camera started running shortly before the deformation and stopped right after the deformation so that the whole deformation process was recorded. The results were analysed by an L & W Analyst projector. Films can be shown with regular movie speed (24 frames sec^{-1}) or slower. Pictures were printed from the films.

2.3. Compression moulding of polymer composite

In order to study the effect of rubber particles on the propagation of shear bands, a plate of high impact polystyrene (HIPS) obtained from the Dow Chemical Company was compression moulded between two blocks of PS. A moulding press consisting of a Buehler specimen mount press and a mould assembly was used. Two discs of PS about 10 mm thick and a disc of HIPS about 1 to 2 mm thick were made separately by using the mount press. The piece of HIPS was then sandwiched between the two PS blocks and they were moulded together using the same mould assembly. The moulding procedure was that the materials were first compressed under a pressure of 41.4 MPa in the mount press. The pressure was maintained until the materials inside the mould were softened and homogenized by heating. After the materials inside the mould were completely softened, as indicated by the stability of pressure, the mould was continuously heated for 10 more minutes and was cooled slowly to room temperature. Specimens were then mechanically and thermally treated again as was described in Section 2.1. The rubber (70–30 butadiene–styrene co-polymer) content in the high impact polystyrene was about 5 vol% and it was distributed uniformly in the PS matrix in the form of spherical particles with sizes in the range of 2 to 6 μm, as will be shown later.

2.4. Scanning electron microscopy

Specimens were coated with a thin layer of gold by using a Technics HUMMER II sputtering apparatus at a voltage of 10 V and a sputtering period of 4 min, and were then examined under a Coates and Welter model HPS–70B field emission scanning electron microscope (SEM) with a tilt angle of about 45°. Pictures with large depth of focus field were taken.

3. Experimental results

3.1. Propagation of a shear band packet without obstacles

The rates of growth of coarse shear bands are usually very high. To study the growth behaviour in detail, the technique of high speed cinematography was employed. The details of this technique have been described in the experimental section. By varying the lens system of the high speed camera, movies of different magnifications can be taken. Fig. 1 is a sequence of pictures which shows the development of a packet of shear bands from a hole. The original series was taken at a speed of 6000 frames sec^{-1}. However, only a few typical stages illustrating obvious developments are shown here. It is seen that during deformation shear bands were initiated from the hole and propagated away from it along straight paths. The coarse bands become more well defined as they grew longer.

A detailed examination of the initial stages under a darkroom enlarger revealed that each single coarse band did propagate during deformation, at

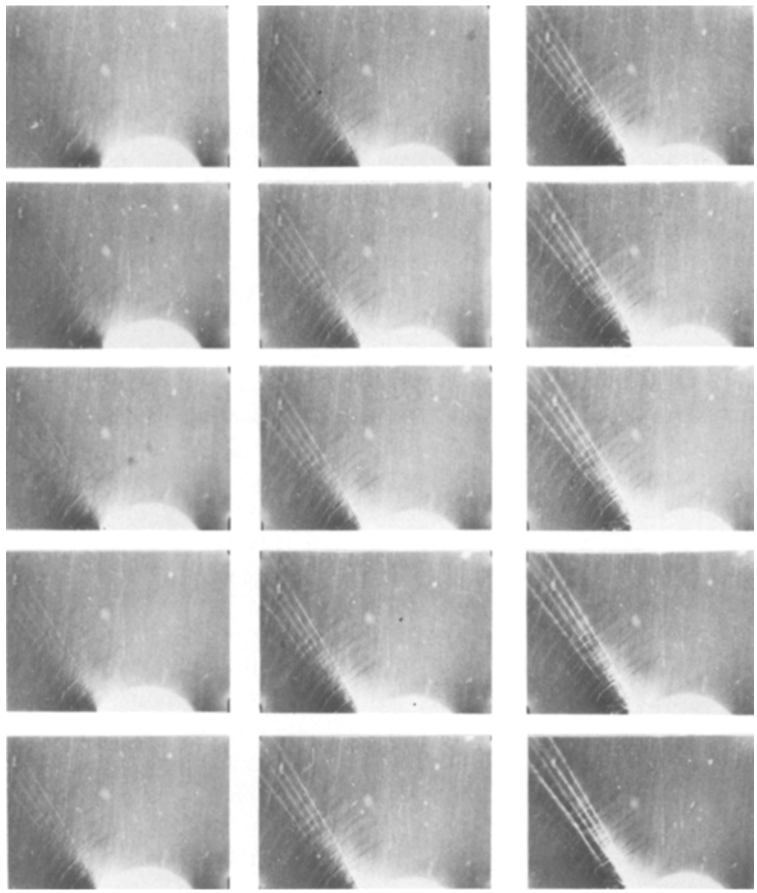

Figure 1 High speed movie sequence showing the development of a packet of shear bands from a hole.

least within the period of observation. Based on these consecutive movie sequences, the lengths of several resolvable coarse bands are plotted against time as shown in Fig. 2. The slope of the plot gives the velocity of the shear bands and ranges from 23 to 185 mm sec^{-1} except for small fluctuations. An average is about 80 mm sec^{-1}.

As the specimen was deformed further, the coarse shear bands within the packet increased in number and the whole packet of shear bands appeared to become a thick band in the later stages of deformation, as shown in Fig. 3. Finally the specimen fractured along the thick band, a process which was reported previously [5]. Several compressions using the same strain rate ($\dot{\epsilon} \approx 0.12\,sec^{-1}$) were made and they all behaved similarly. It appears that when the coarse shear bands within a packet developed to a certain extent, a large scale shear motion occurred inside the packet between the existing coarse shear bands. Through this large scale motion, the band packet became a thick band with a very localized shear region and a distinct measurable shear strain as reported before [3]. A detailed examination showed that when the second-stage shear took place, it seemed to occur simultaneously along the entire band packet with a measurable shear rate. A movie sequence showing this second shear is shown in Fig. 4 revealing the development of a thick shear band from a small hole. It is seen that, as compression continued, the thin shear bands became more and more abundant and when the process reached a certain limit, the second-stage shear motion occurred, as seen by the step formed at the hole. The thickness of the band packet, which later became the thick band, seemed to have remained the same during the whole shearing process.

The height of the step was measured and is plotted as a function of time, as shown in Fig. 5. It is seen that the shear rate seemed to be highest at the beginning of the motion, except for some incubation period during the production of coarse bands before the large scale shear motion. The shear rate gradually decreased to zero during

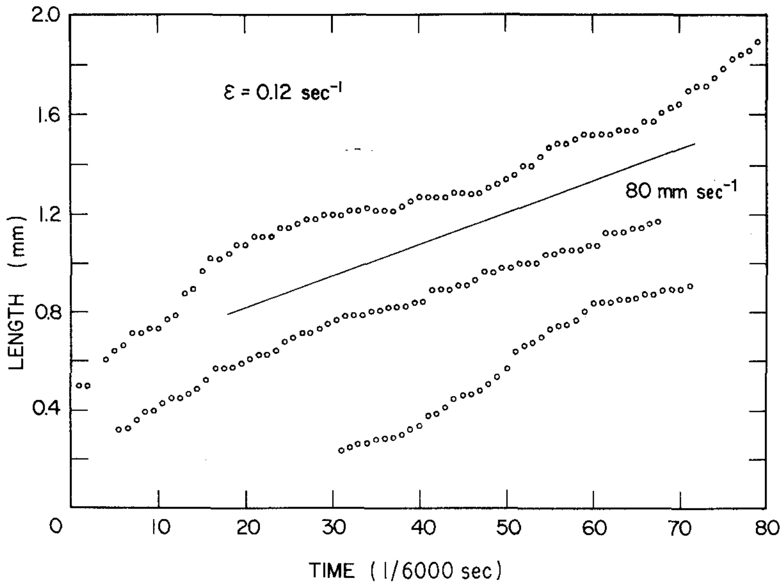

Figure 2 Increase of the lengths of coarse shear bands during propagation.

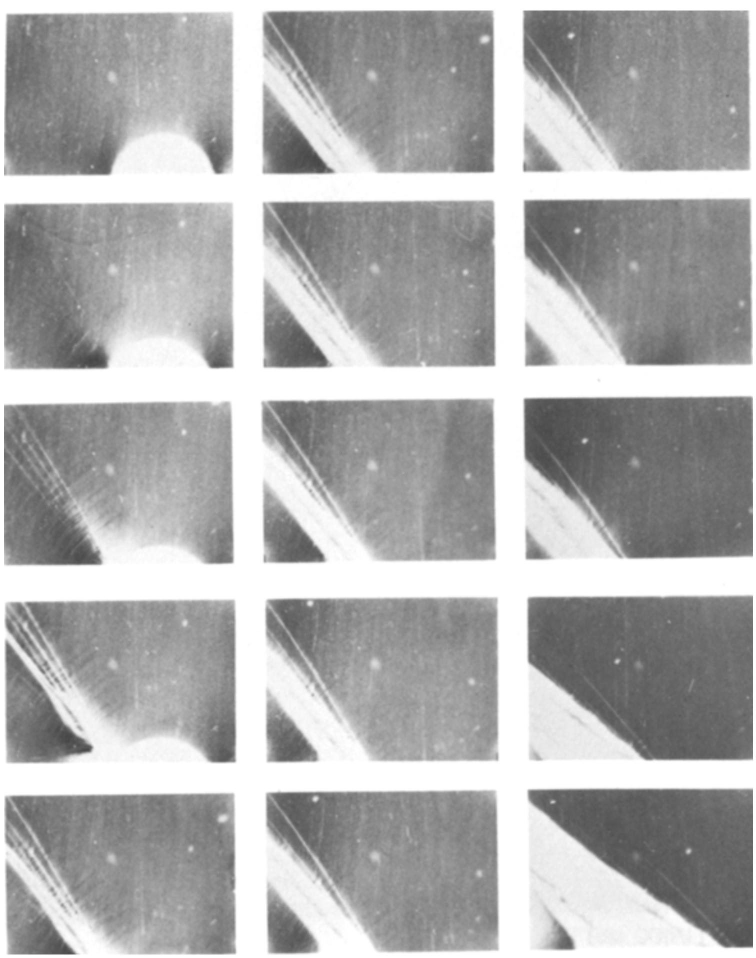

Figure 3 High speed movie sequence showing the formation of a thick band within the coarse band packet.

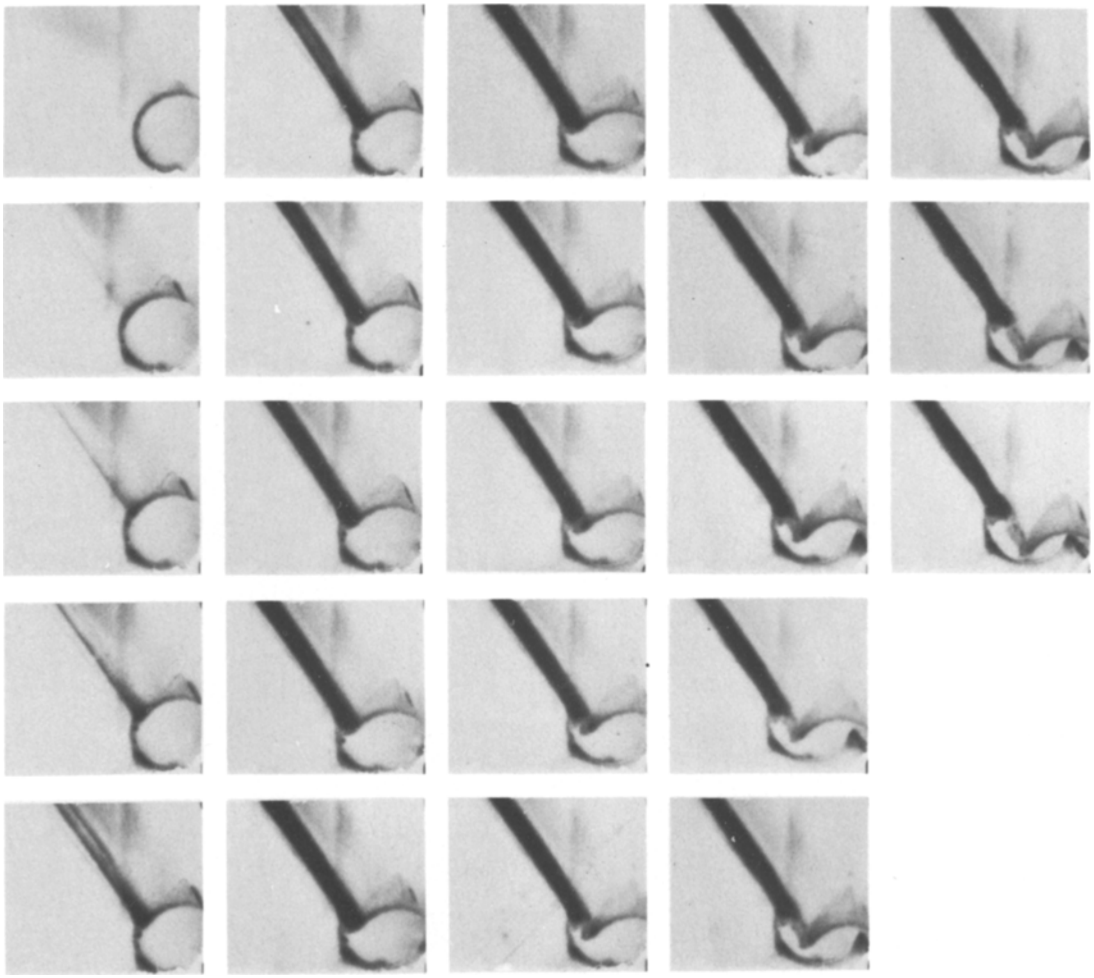

Figure 4 High speed movie sequence showing the development of a thick band by a large scale shear motion.

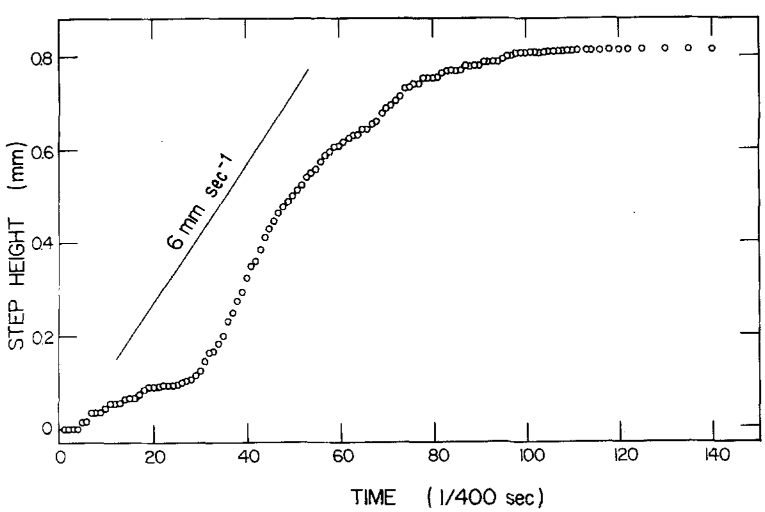

Figure 5 Step height at the hole as a function of time during the development of a thick band. Fracture was avoided by unloading near the end.

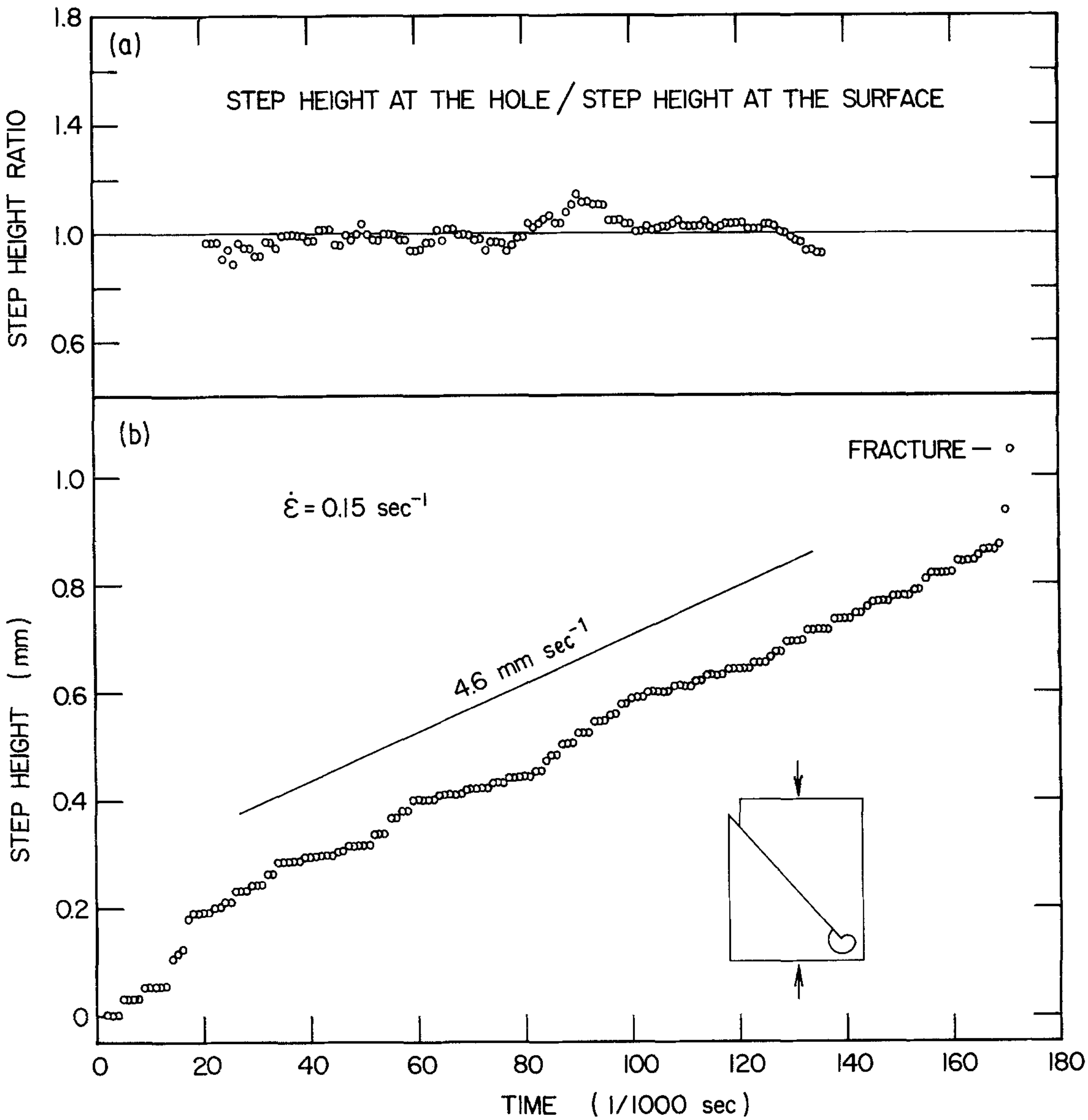

Figure 6 (a) Step height ratio and (b) step height at the surface as a function of time during the development of a thick band followed by shear fracture without unloading.

unloading. The average shear rate as measured by the slope is about 6 mm sec^{-1}, corresponding to a shear strain rate of 24 sec^{-1}. To see the behaviour without unloading, another movie sequence was taken and the results are shown in Fig. 6. It is seen that the shear rate remained almost constant at 4.6 mm sec^{-1} (corresponding to a shear strain rate of 19 sec^{-1}) until shortly before fracture. About a millisecond before separation a sudden increase in shear rate is seen to take place. This is attributed to shear at the interfaces between the thick band and the undeformed material. At such interfaces some fibres were seen to be stretched to fracture. Upon annealing, these stretched fibres were fractured rather than shrinking. These will be discussed in a later communication. Some results on the fracture of shear bands have been reported already [5]. The ratio of the step heights at both ends of the band during large-scale shear motion as measured on each frame is also plotted in Fig. 6. It is seen that the average ratio is about 1, indicating that the large-scale shear motion happened almost simultaneously along the entire band.

3.2. Propagation of a shear band through an existing band

3.2.1. Surface morphology and topography

In a previous paper [4], it is shown that intersections of coarse shear bands always caused mutual shearing as seen under the electron microscope. This effect was attributed to alignment of the molecular chains inside the coarse bands and the easier shear along molecular directions.

As in the case of thin coarse bands [4], to produce the first type of intersection, a notched specimen was deformed first to produce a thick band, as shown in Fig. 7a. A Dektak trace (see Fig. 7b) shows that the h/b (height-to-base) ratio is about 0.087; many such traces were made along the band and were recorded. Another notch was then made at the other side of the specimen, and the specimen was compressed again in the same compression direction to produce the intersection. Fig. 8a shows the same band after the intersection. It is seen that the old thick band was sheared by the new band. The new intersecting band is shown in Fig. 8b. Away from the intersection it was a well-defined thick band. As it approached the intersection, the main propagation direction was changed; there was also splitting of the band or the generation of new coarse bands at the intersections. This observation seems to indicate that the second-stage large-scale shear motion of the new band packet was not completed owing to the presence of the existing thick band. The intersection mechanism, however, can still be understood by the same proposed process [4].

To see how the surface profile changes, a Dektak trace was made at the intersection (see Fig. 8c). The trace direction made an angle with both bands and is indicated by the arrow in Fig. 8a. The height of the old band at the intersection was greatly increased after being intersected by the new band. The new height was 76 μm, more than twice as high as the 31.4 μm observed before the intersection at the same location. However, the h/b ratio, 0.11 as measured from the Dektak trace, was comparable to that of 0.087 before the intersection.

To see the propagation of the new band in more detail, the old thick band was polished away so that only microcracks oriented more or less along the fibre directions were left behind. The intersecting new band packet is shown in Fig. 9. It is seen that the new shear bands tend to change their directions of propagation toward the directions of microcracks. A group of thin bands changed their directions even before meeting the old band and propagated between the microcracks. The resisting effect of the old band must be sufficiently large since the group of thin bands did not even get through the old band, while the main band did get through.

3.2.2. Propagation speed

To see the effect of an existing shear band on the rate of propagation of a new band, the technique of high speed cinematography was employed again. Fig. 10 shows the developing stages of shear bands from a hole with an existing thick band nearby. The original movie was taken at a speed of 2000 frames sec^{-1}. It is seen that in this specific case, the band packet near the existing band developed first. However, when it reached the existing band the propagation seemed to be stopped, with only a few thin bands getting through. In the meantime, the other band packet from the hole began to develop fully and quickly became a thick band. Then more thin bands got through the intersection and finally the large-scale shearing process took place to transform the intersecting band into a thick band. The existing thick band was then sheared by the intersecting band during this process.

The length of both band packets during propagation was plotted against time as shown in Fig. 11. The velocity calculated for the free propagating band packet is about 44 mm sec^{-1} while that for the intersecting band packet showed a considerable delay at the intersection. The existing band took about 30 milliseconds to allow the new band packet to pass through, which corresponds to a propagation speed of about 17 mm sec^{-1} as shown on the plot. This speed is much slower than that of the free propagating band. However, after crossing the existing band the propagating band seemed to resume the speed for free propagation.

Fig. 12 shows another movie sequence selected from the original strip taken at 500 frames sec^{-1}. The shear displacement at the hole was measured and plotted against time in Fig. 13. The displacement rate of 2 mm sec^{-1} corresponds to a shear strain rate of 10.5 sec^{-1} inside the shear band. As pointed out earlier, the slowing down of displacement rate was due to unloading. Without unloading, the shear rate will remain constant up to fracture.

3.3. Effect of fully recovered bands

3.3.1. Morphology of intersection

To see if a fully recovered band can still affect the

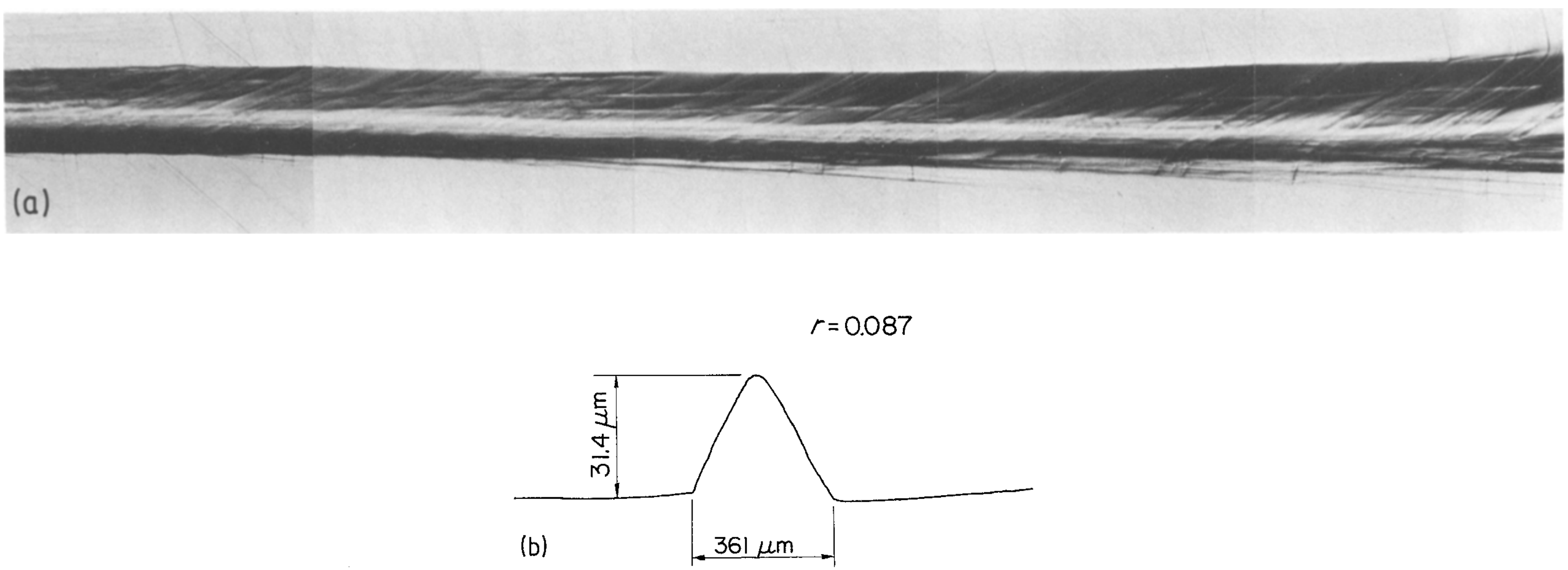

Figure 7 A thick band produced from a notch after compression. (a) Photograph (b) Dektak trace.

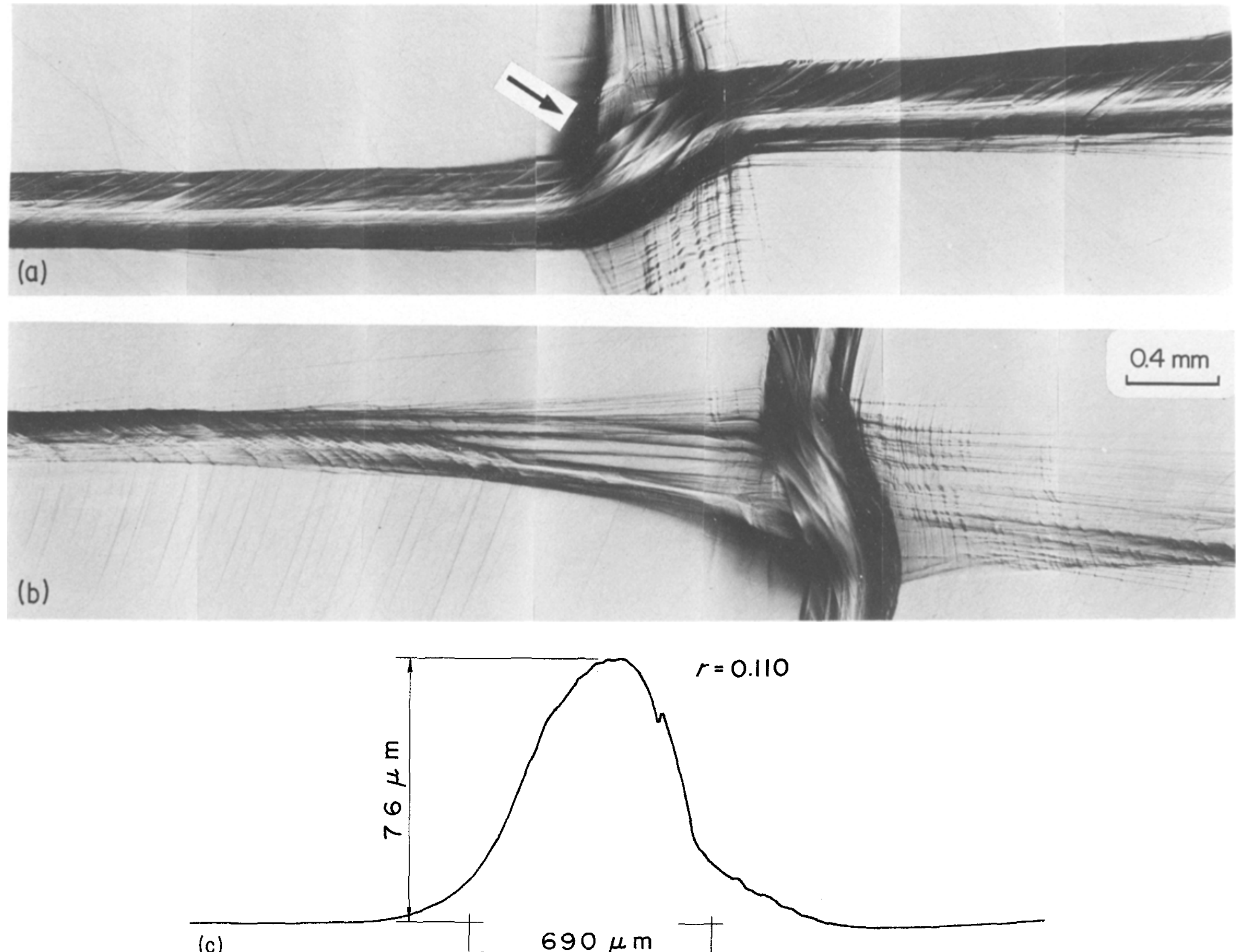

Figure 8 The thick band of Fig. 7 after being intersected by a new band, showing (a) Photography of the old band, (b) Morphology of the new intersecting band, and (c) Dektak trace of the intersection, taken at the angle indicated by the arrow in 8a.

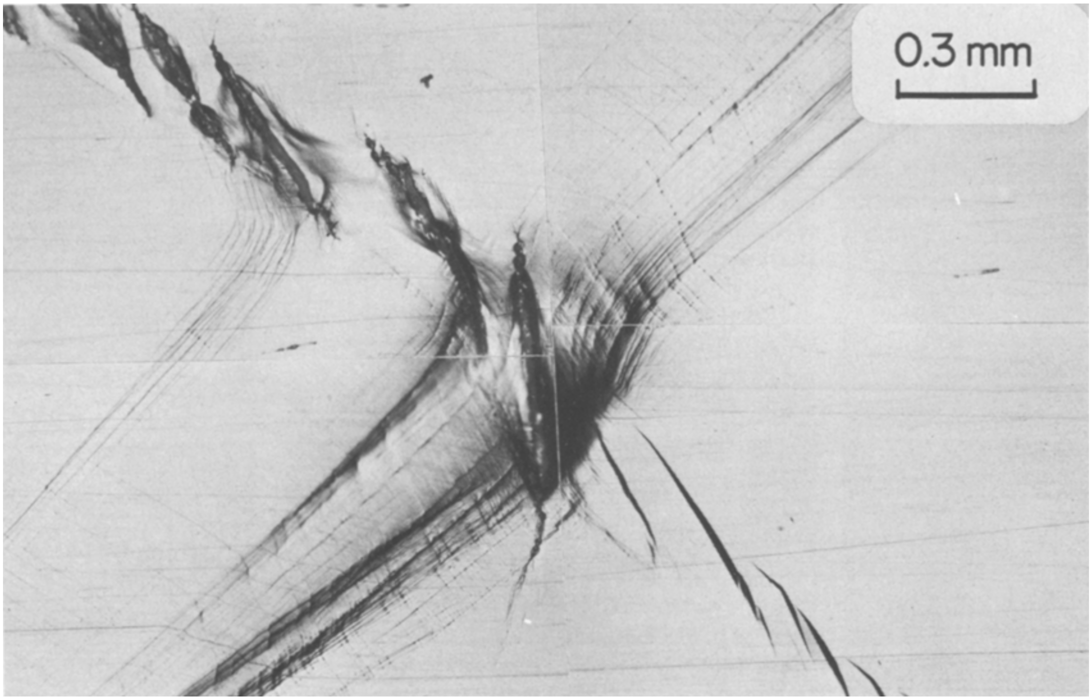

Figure 9 The intersection of a new band packet with a polished old thick band showing the change of the orientation of the microcracks at the intersection inside the old band. (OM)

propagation of an intersecting band packet, an experiment involving the intersection between a propagating band and a fully recovered band was made. Since shear bands usually leave no traces after being fully recovered, a gold decoration technique as used previously [1] was employed. A thick banded specimen was first coated with a thin layer of gold on the banded surface. The specimen was then annealed at 110° C for 3 h. The thick band was fully recovered as evidenced by the straightening of the scratches on the surface. The band locations, however, were revealed by the collapsing of the gold on the thick band. Fig. 14 shows the intersection of a new band packet with the recovered band. It is seen that the propagation of the new band packet appears unaffected by the recovered band. While the recovered band was sheared, the propagating band remained straight when passing through the recovered band. The mutual shearing effect for the intersection of unrecovered bands as reported before [4] does not exist in this case. The material inside the recovered band seems to have returned to its original undeformed state.

3.3.2. Speed of propagation

To see if the velocity of propagation of the new band packet was also unaffected by the intersection of the recovered band, a thick-banded specimen was first polished to remove the band and annealed at 110° C for 3 h. The thick band was fully recovered. However, as reported in a previous study [3], the old band location developed a valley due to the polishing of the band before annealing. This valley showed up clearly under the high speed movie camera so that the propagation of the new band through the recovered band could be seen. The length of the band packet was plotted against time and is shown in Fig. 15. It is seen that the recovered band did not seem to have any effect on the propagation speed of the new band packet.

3.4. Effect of rubber particles

3.4.1. Speed of propagation

To see the effect of some other obstacles such as butadiene–styrene rubber particles on the propagation of shear bands, a high impact polystyrene disc containing rubber particles was compression moulded between two PS discs as mentioned in the experimental section. A small hole was drilled at one corner in the PS region. Upon compression at a high strain rate, shear bands were initiated from the small hole and propagated across the rubber-

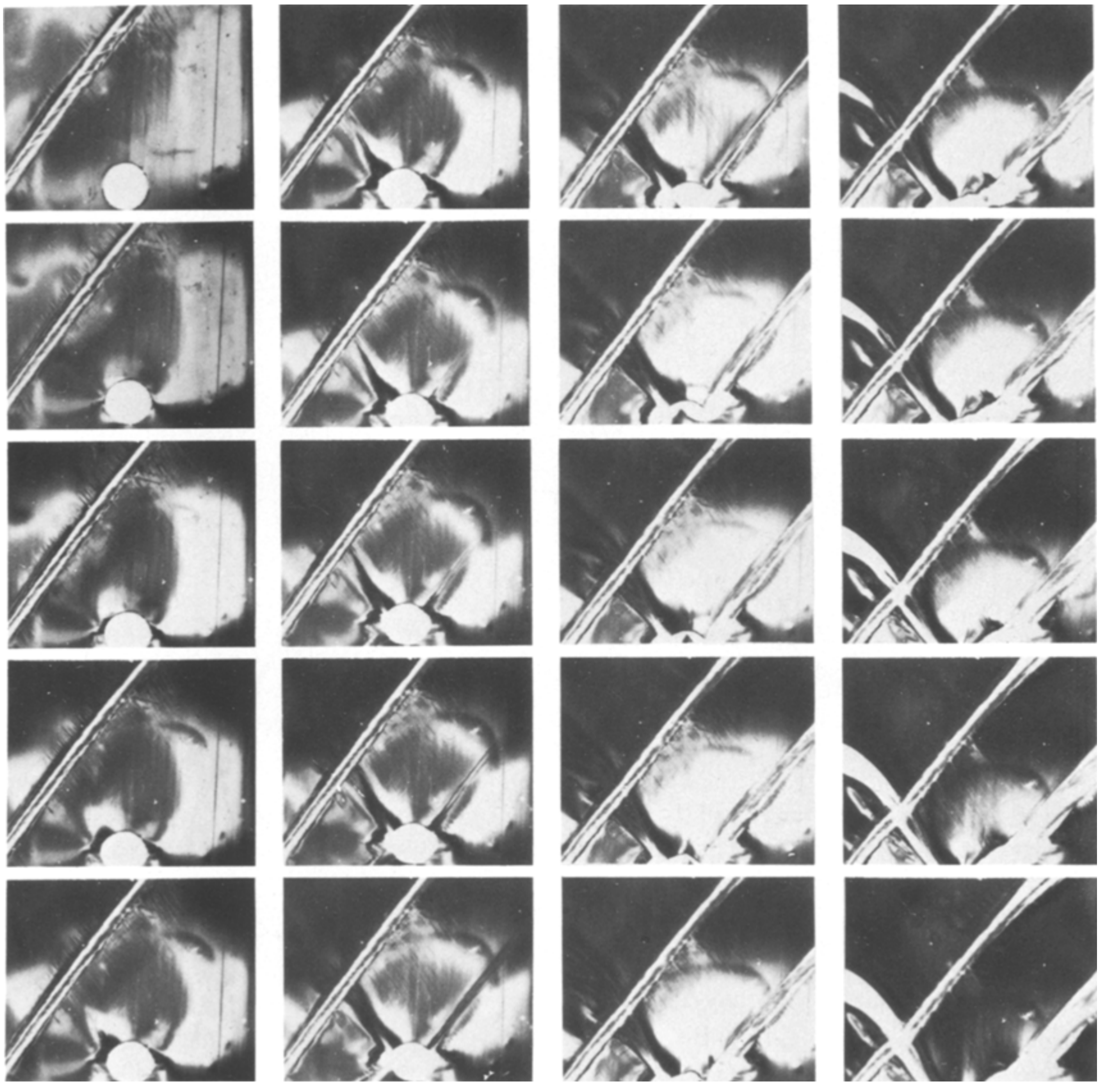

Figure 10 High speed movie sequence showing the development of shear bands from a hole toward and across an existing thick band.

modified region towards the side surface of the specimen. Again, the propagation process was studied by using high speed photography. A set of movie sequences are shown in Fig. 16 which were selected from the original strip taken at a speed of 1000 frames sec^{-1}. It is seen that during compression the dark rubber-modified region resisted the propagation of the band packet, which became dispersed after crossing the region. The length of the band packet is shown as a function of time in Fig. 17. It is seen that the propagation of the band packet slowed down even before approaching the rubber-modified region. The velocity of the band packet before the intersection was about 145 mm sec^{-1} as shown. The average speed in the intersection was about 56 mm sec^{-1}. However, the band packet picked up the speed as soon as it passed the rubber-modified region. It is obvious that the rubber particles provide sufficient resistance for the propagation of shear bands in polystyrene.

3.4.2. Optical and electron microscopy

To see in more detail how these rubber particles resist the propagation of shear bands, specimens were examined under an optical microscope. An example is shown in Fig. 18. It is obvious that the shear band dispersed and split into many thin bands even before appraoching the rubber-modified

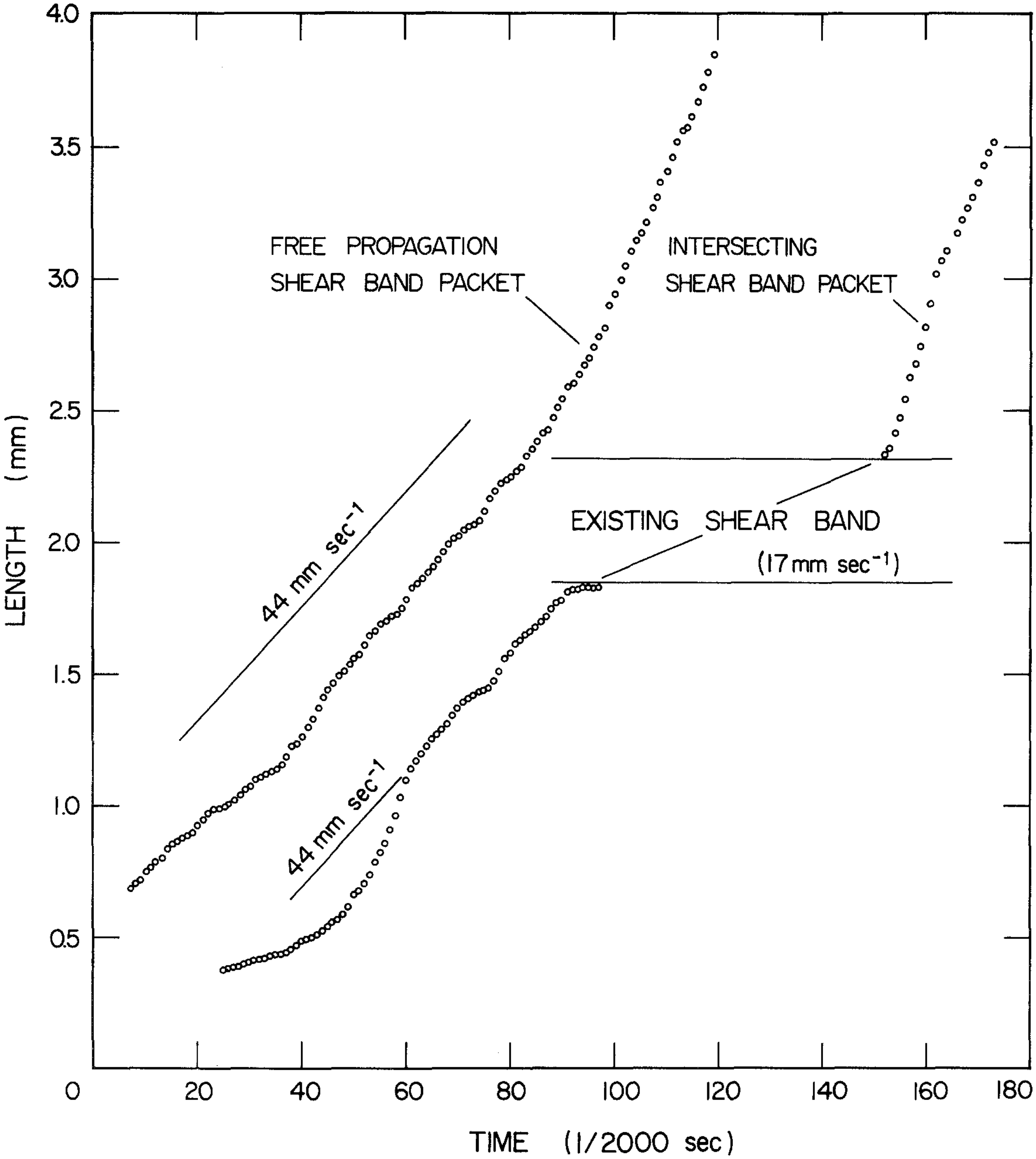

Figure 11 The lengths of shear band packets during propagation with and without an existing thick band in the path.

region. The shear strain in the band decreased at the same time showing that the large-scale shear motion as discussed earlier did not take place uniformly along the whole band. The thin bands dispersed even more inside the rubber-modified region but managed to reappear after passing through the region. Such reappearance could be a result of joining of the fine bands dispersed between the rubber particles.

Some details of the joining process are shown in Fig. 19. The fine bands in the rubber-modified region seem to combine into thin bands outside the region and propagate in that mode. At a magnification of about 450 times the fine bands appear to disperse themselves uniformly inside the rubber-modified region. However, at a magnification of 4000 times, the fine bands are seen to cluster together into bundles as shown in Fig. 20. These bundles seem to lie between the rubber particles although some were in the particles also.

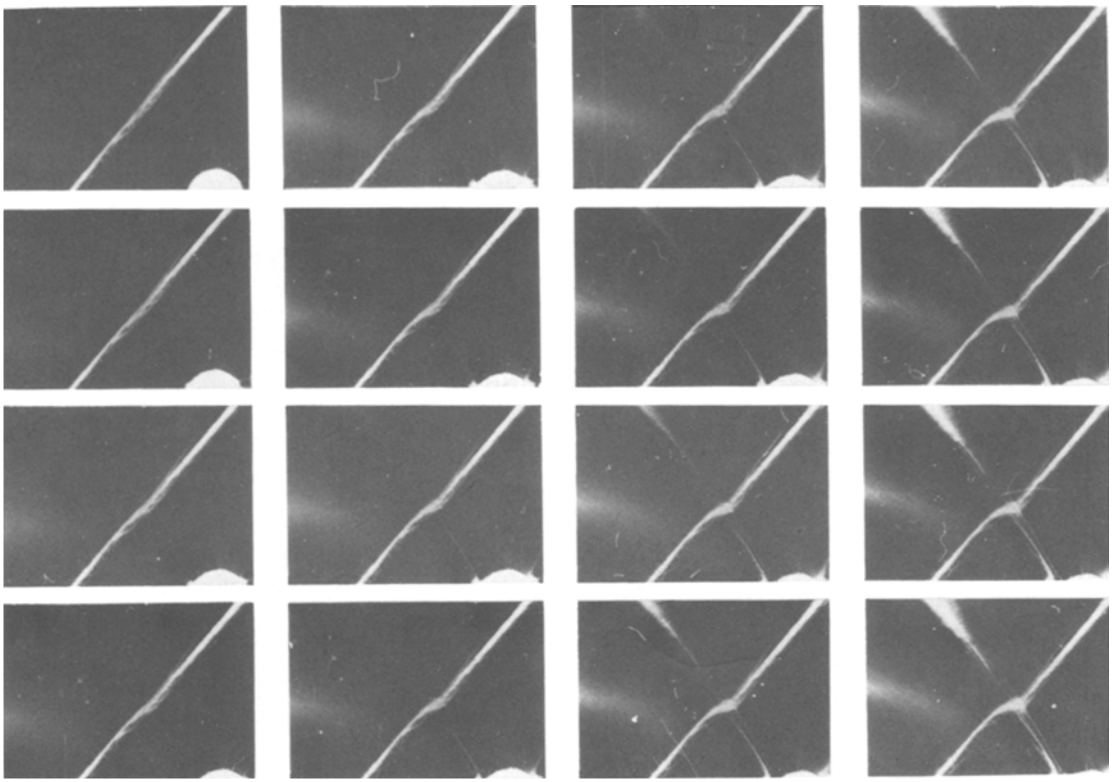

Figure 12 Another example of a high speed movie sequence showing the intersection of a propagating band packet with an existing thick band.

It is seen that the rubber particles are very effective in dispersing slip so that the only mode of deformation is the spreading of fine bands. This capability prevents not only the formation of thin bands but also the clustering of them which is a necessary step before the large-scale shear motion during the formation of thick bands. As shown before [1, 2], the fine bands are a ductile mode of deformation. The ability to prevent the formation of thin and thick coarse shear bands must have contributed to the superior mechanical properties of HIPS [7–11].

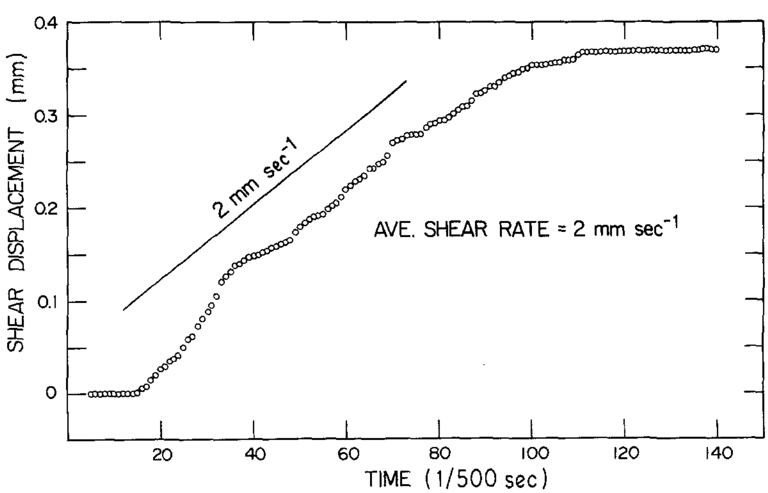

Figure 13 Shear displacements of the existing band during intersection by a new band packet.

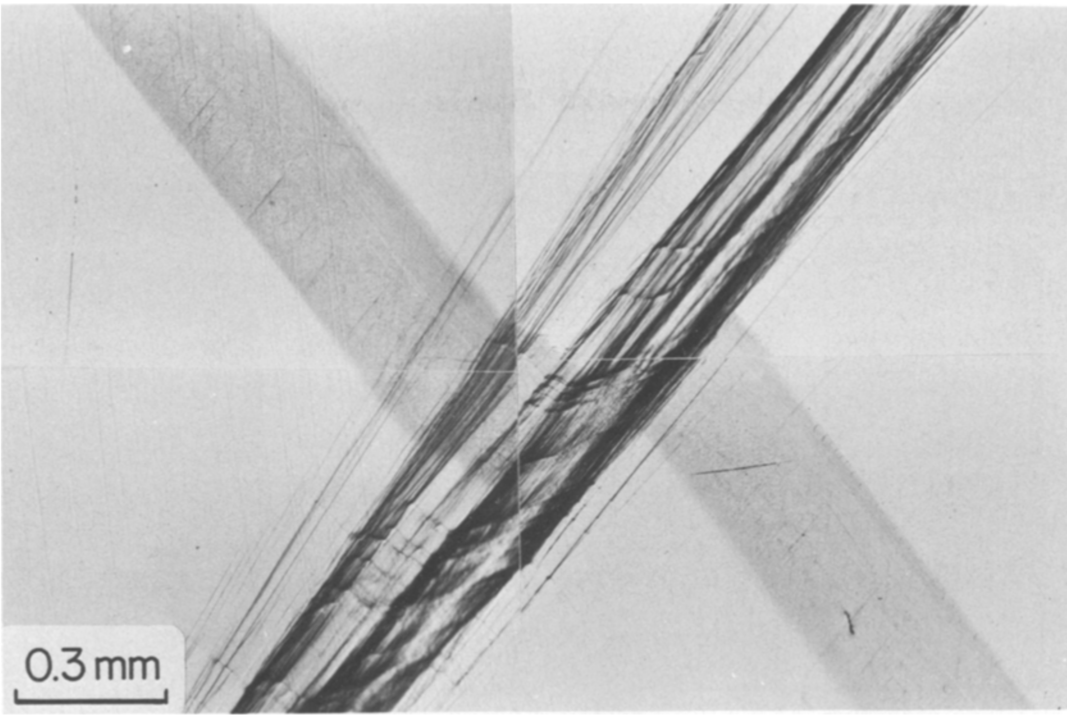

Figure 14 Optical micrograph showing the intersection of a new band packet with a recovered thick band revealed by collapsing of gold particles over the old band.

4. Remarks on shear mechanisms

The behaviour reported here has some similarity with that of crystalline substances. For example, the slip lines in metals also grow and propagate, as reported in aluminium by Chen and Pond [12] and by Becker and Haasen [13]. The speed of propagation was not uniform either. Some distribution of such speeds was reported more recently by Pond [14] with data on Cu and α-brass. While the smallest speed could be zero, the largest speeds observed were about 0.4 to 4 μm sec^{-1}. However, speeds as high as 10 to 70 μm sec^{-1} were observed by Becker and Haasen. Instead of slip lines, the velocity of dislocations observed by using etch pits

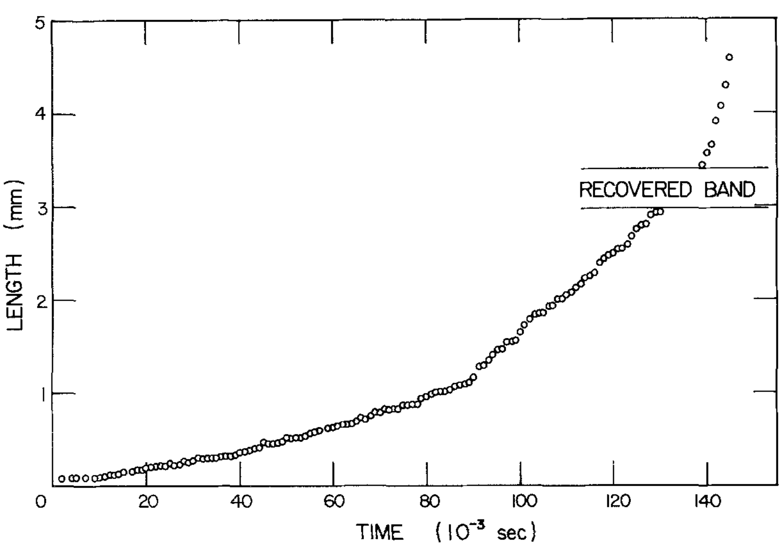

Figure 15 Lengths of a band packet as a function of time during the intersection of a recovered band.

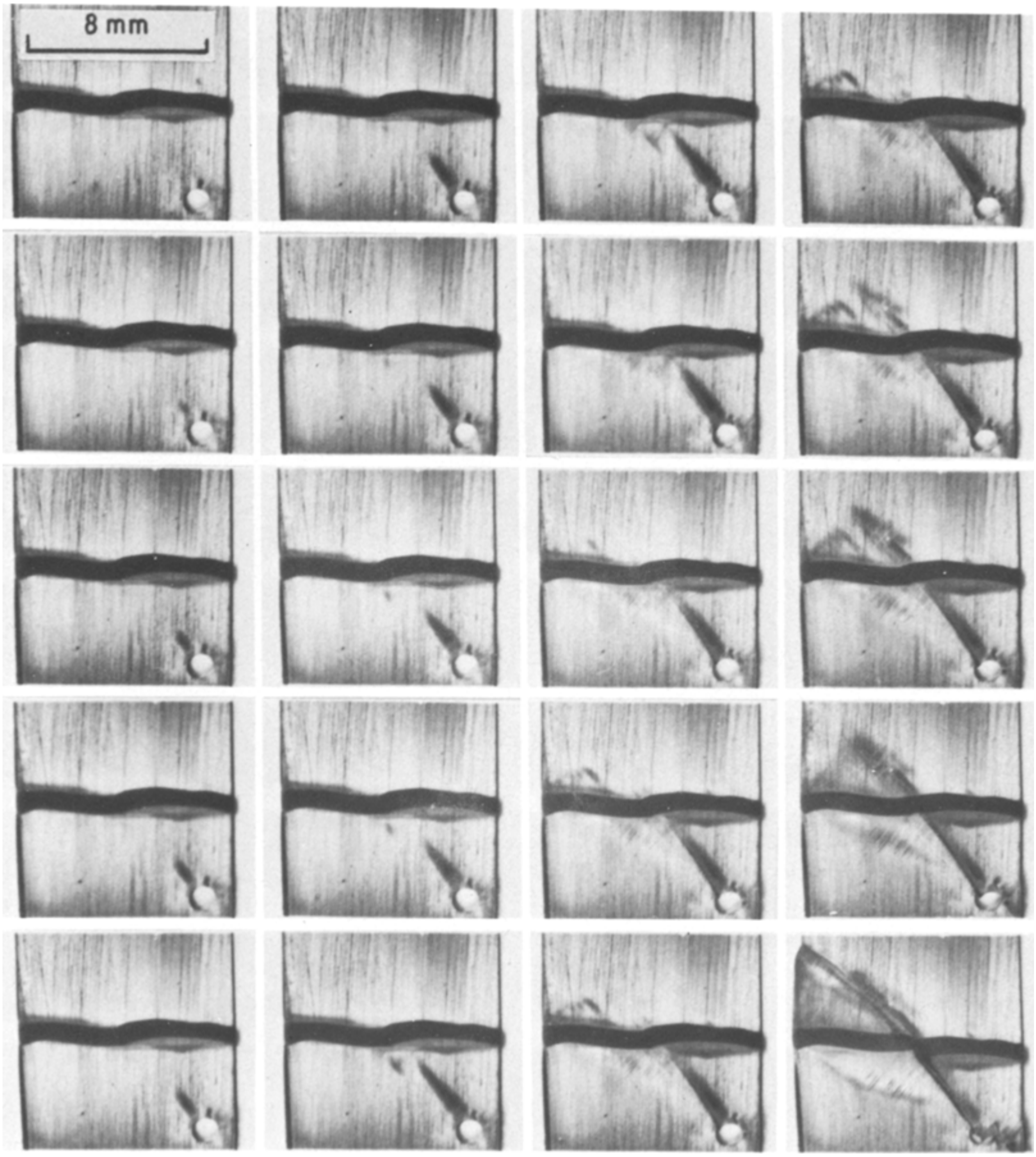

Figure 16 High speed movie sequence showing the propagation of a shear band packet through a thin layer of rubber-modified region.

ranged from 10^{-5} to 10^{4} cm sec^{-1} [15]. The speed observed here in PS, 10 to 20 cm sec^{-1}, is sufficiently high, suggesting collective motion of atomic groups or molecules during the propagation of shear bands.

Such collective motion which travels by a front, like a dislocation, can interact with another front travelling in a different direction. An easy way of illustrating such interaction is by the use of dislocations [16]. The interaction produces a new front which may be immobile such as the Lomer–Cotterell lock. These immobile or slow-moving fronts could be used to explain the observation that new shear bands are slowed down by existing shear bands. The propagation inside the existing shear band takes place by the motion of a slow-moving new front produced by the interaction of the two shear bands.

Similar to the behaviour of dislocations, the shear fronts can change their directions of propagation upon meeting obstacles such as the rubber particles in HIPS and thereby disperse themselves. Some early examples of fine slip observed in precipitation-hardened Al are reported by Carlsen and

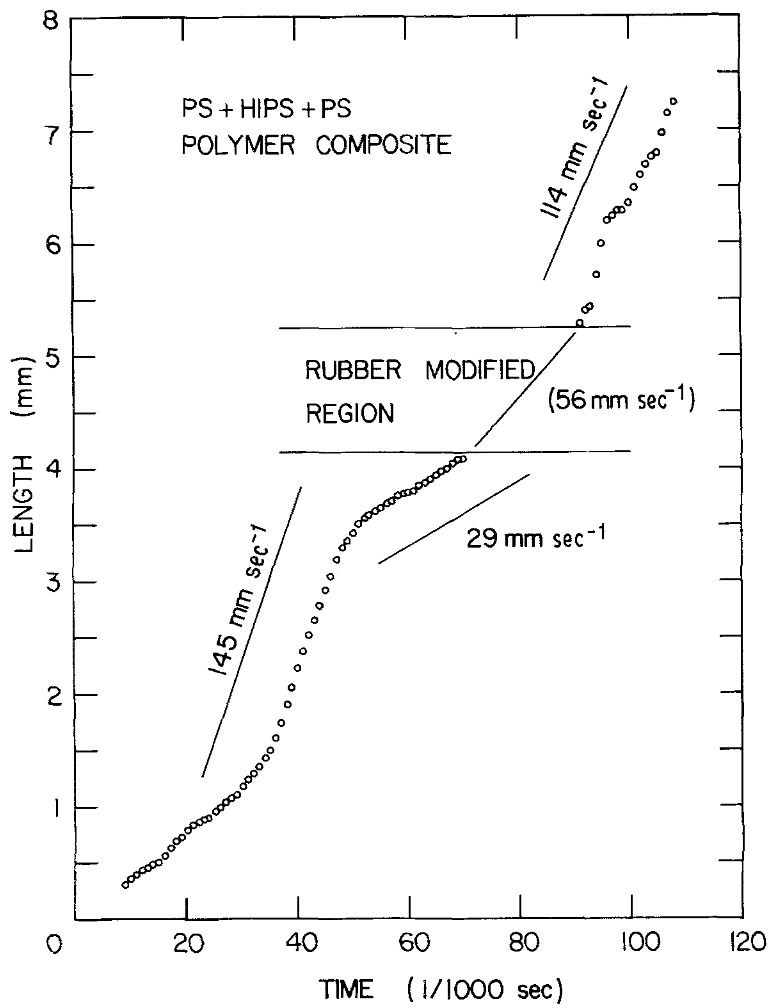

Figure 17 The lengths of the shear band packet during propagation across the rubber-modified region.

Honeycombe [17], Greetham and Honeycombe [18] and Thomas and Nutting [19]. While the slip lines are coarse and straight in supersaturated solid solution of Al–4.5 wt% Cu alloy (no precipitates) deformed at 77 K, they are fine and wavy in aged alloys (with precipitates). Such change of slip patterns was explained by Hirsch [20] using the cross-slip property of screw dislocations. A similar explanation can be advanced here, since the concept of a propagating shear front seems to have been established by the observations reported in this communication.

5. Summary and conclusions

When atactic PS was compressed at a strain rate of $0.12\,sec^{-1}$, a shear band packet consisting of thin coarse bands developed at stress concentrations. A single resolvable coarse band was observed to propagate at between 23 and 185 mm sec^{-1} with an average speed of 80 mm sec^{-1}. Then, the number of thin coarse bands in the packet became so numerous that the packet became a thick band by a large scale shear motion, which seemed to take place throughout the packet. Such shear motion had an average shear rate of about $6\,mm\,sec^{-1}$, corresponding to a shear strain rate of $24\,sec^{-1}$.

When a propagating coarse band packet met an existing shear band, the packet dispersed itself into thinner bands and spread out to cover more spaces. At the same time the rate of propagation slowed down to about one half of the original speed. Individual thin coarse bands seemed to propagate in the fibre or microcrack directions in the existing band. After the thin coarse bands passed through the existing band, they joined together to form a thick packet which resumed the high speed of propagation. When the large-scale shear motion took place, the shear displacement was about the

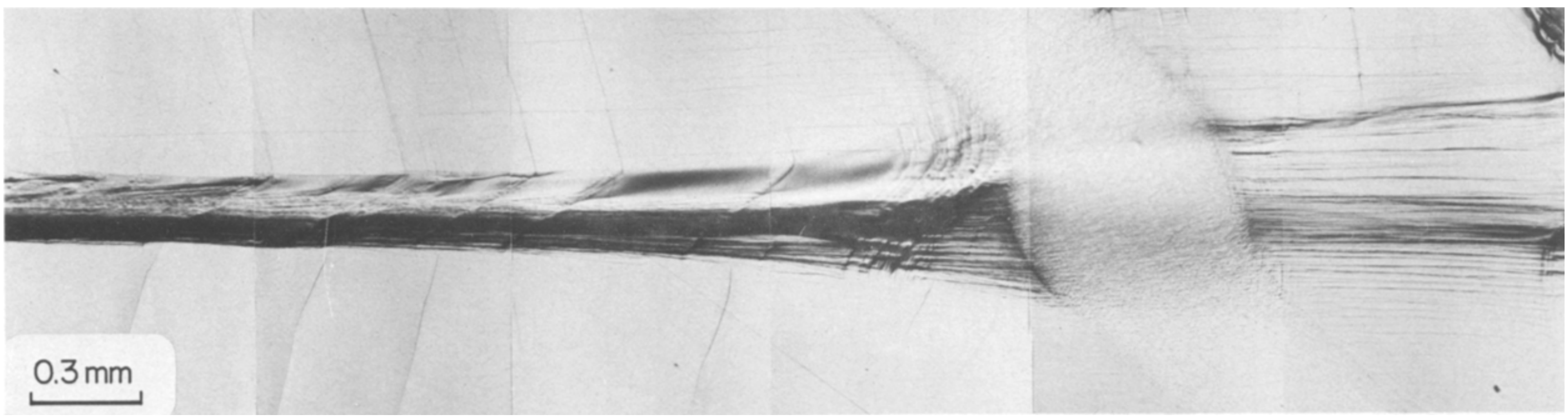

Figure 18 Optical micrograph showing the morphology of the intersection of a thick band with a thin layer of rubber-modified region.

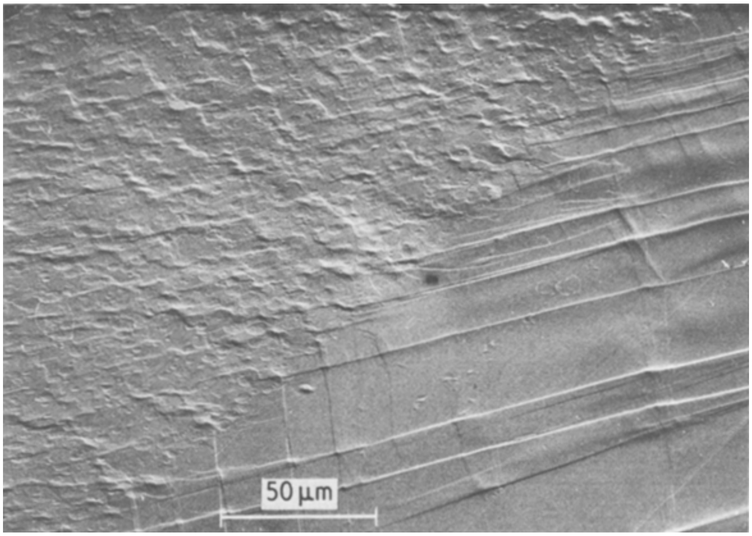

Figure 19 SEM details of the joining of the dispersed shear bands emerged from the rubber-modified region into thin coarse bands.

same throughout the packet, but the shear strain was much smaller at and near the interaction. The displacement rate, $2\,\mathrm{mm\,sec^{-1}}$, was also smaller than the case without the existing band as an obstacle ($6\,\mathrm{mm\,sec^{-1}}$).

A fully recovered shear band did not offer any observable resistance to the propagation of a new band packet, either in surface morphology or in the speed of propagation.

A region of HIPS which had a dispersion of 5% 2 to 6 μm rubber particles did offer considerable resistance to the propagation of a shear band packet. First, the packet slowed down to about 1/5 of its speed even before appraoching the HIPS region. This reduced speed caused spreading of the thin hands. Secondly, the thin bands "disappeared" into the HIPS region or they were transformed into discontinuous fine bands which sometimes clustered between the rubber particles. These fine bands recombined into thin bands which joined and gathered together into a band packet outside the HIPS region and resumed the speed of propagation.

These observations established the concept of a shear front in the propagation of shear bands which enables us to use the properties of dislocations to understand the behaviour of propagating shear bands in meeting obstacles such as an existing shear band and a HIPS region.

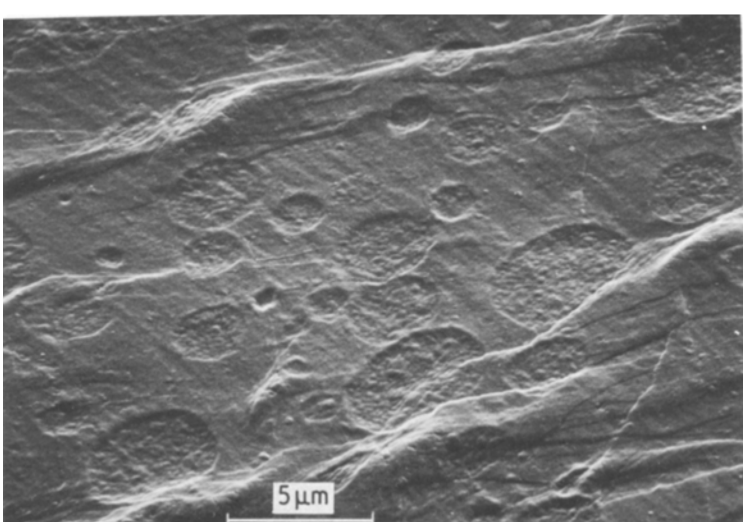

Figure 20 SEM showing bundles of fine bands dispersed between and inside rubber particles.

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