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Slip Deformation in Drawn Polyethylene Films

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Synopsis

When well oriented and crystallized high density polyethylene film was redrawn, slip deformation and twin deformation, the mode of which depends upon the redrawing angle to the original drawn structure, could be observed. When the redrawing angle is very small, single slips, the direction of which is [001] to the original drawn structure, and homogeneous deformation bands attached to them are usually observed. The [001] directions of slipped and unslipped regions are slightly at an angle to each other. As the redrawing angle approaches a right angle, kink bands can be observed. From these results we may conclude that the structure of this drawn polyethylene film is very similar to that of a single crystal or metal.

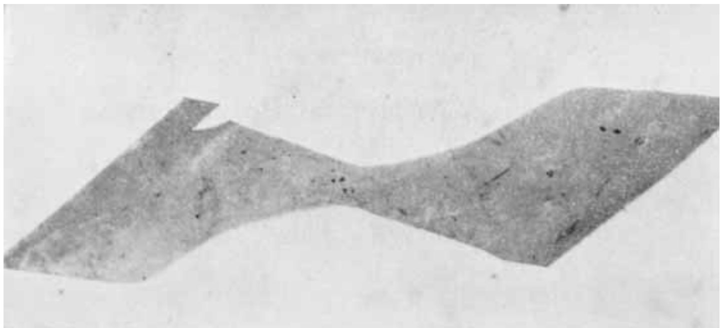
In recent years there have been presented some works[1,2] reporting that the deformations of crystalline polymers can be interpreted in terms of slip and twin deformations, which are the mechanism of plastic deformations in metals. During our studies on the drawing mechanism of polyethylene films, we could observe visually typical sliplike deformations similar to those in metals and therefore there were no doubts about the existence of a metallike deformation caused by the motion of dislocations in a crystal.

EXPERIMENTAL AND RESULTS

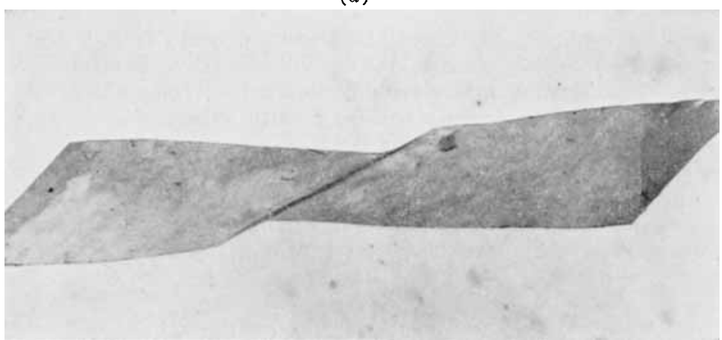
Polyethylene samples used were commercial films of high density polyethylene, of about 0.07 mm. in thickness. Strips of these films were fully drawn at 30°C. and in the boiling water. Strips 5 mm. in width were cut from these drawn films at various angles to the original direction of drawing, redrawn[3] at the rate of about 3 cm./min. at room temperature, and these redrawn films were examined with the optical and electron microscope and by the x-ray diffraction in order to elucidate the deformation mechanism of the polyethylene films. Also stress-strain curves were recorded with an Instron tensile machine type TM, and the results were studied in relation to microscopic observations.

Microscopic Observations

For microscopic observations we used the films originally drawn in the boiling water or the films annealed at about 100°C. to crystallize well after being extended at 30°C. because of the fact that the slip deformations were



(a)



(b)

Fig. 1. Photograph of redrawn polyethylene films. Film was first drawn 800% to the direction of the *a* axis (*a*) at 30° C. and (*b*) in the boiling water, and then redrawn at 45° to the original drawing direction at 20° C. × 2.

observed clearly in these films and were scarcely noticed in the unannealed films drawn at 30°C. Commercial films sometimes showed orientation along the *a* axis, and the direction of the first drawing seemed to have some effect on the occurrence of slip deformations, since the orientation of the crystallites in the drawn film and the uniformity of the structure depend upon the direction of drawing. We chose the *a* axis for the first drawing direction, as the slip deformations seemed to be observed most easily in that case.

Figure 1 shows the redrawn films which were originally drawn 800% in the direction of the *a* axis at 30°C. and in boiling water, and then redrawn at 45° to the original drawing direction at room temperature. We can see the deformation band clearly in the films originally drawn in the boiling water when they are elongated beyond the yield value (about 15% elongation), but the films originally drawn at the lower temperature do not show a deformation band. If a film with slip bands is further extended, slip deformations continue to slip off. This type of deformation occurs easily in the case where the extension ratio of the first drawing is large, the original drawn film is annealed and crystallized well, the rate of redrawing is high, and the angle between the first and second drawing directions is small. When the redrawing angle is intermediate or the rate of redrawing is slow, it does not slip off, but continues to elongate plastically similar to the case of cold drawing, that is, the original drawn and redrawn regions are connected through narrow region and the width of the redrawn structure remains constant during the plastic deformations continue.

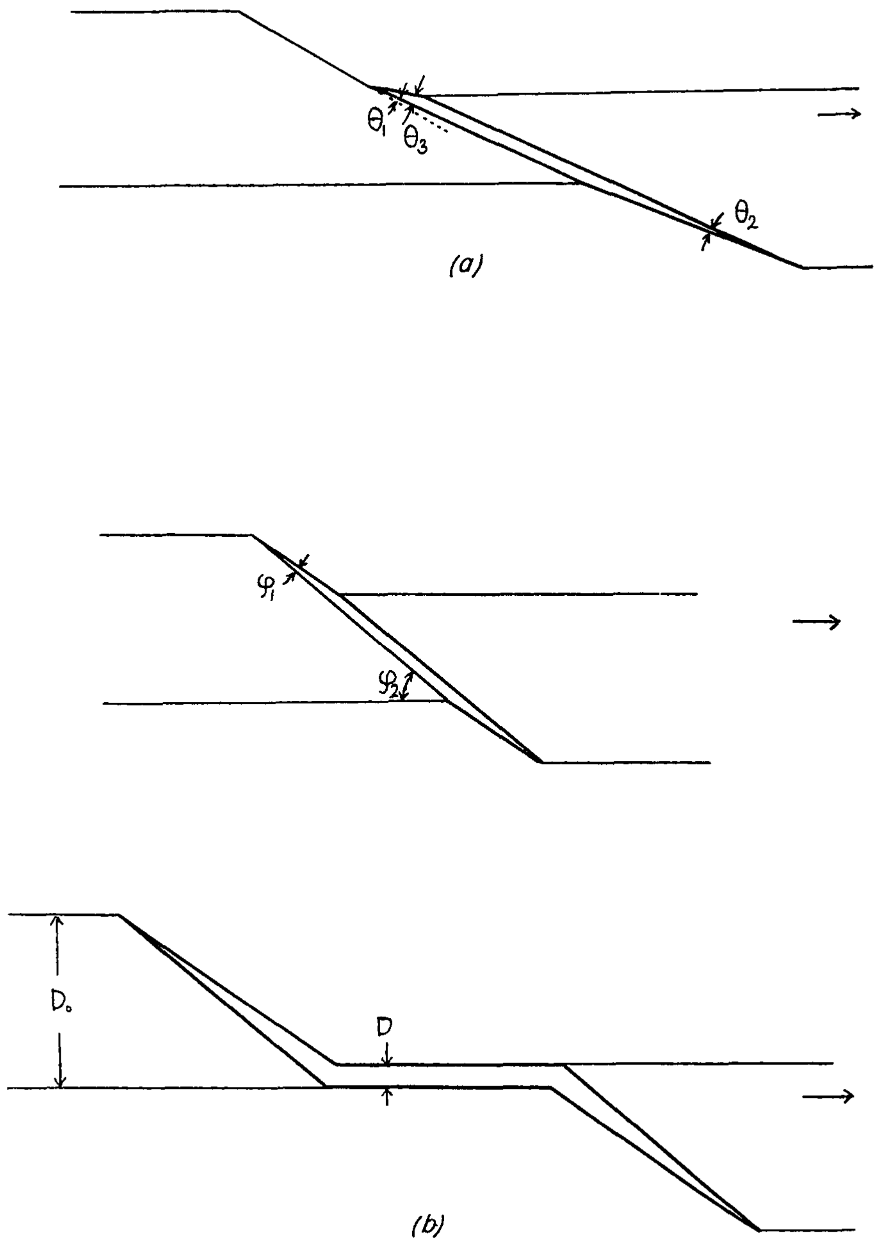


Fig. 2. Schematic representations of two types of deformations: (*a*) the case in which the film slips off; (*b*) the case in which uniform plastic flow occurs. Arrows show the direction of extension.

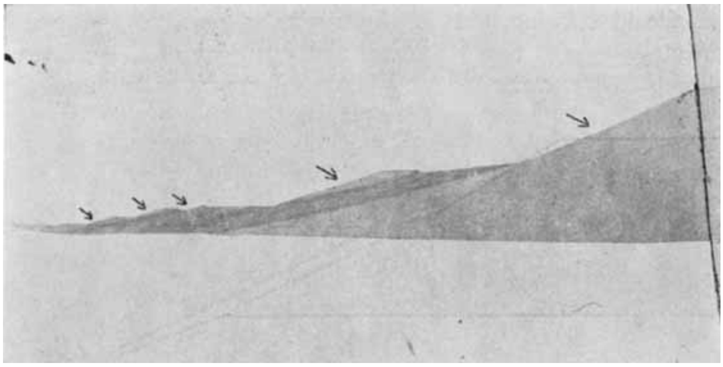


Fig. 3. Photograph of a piece of slipped off polyethylene films. Film was first drawn 1800% to the direction of *a* axis in boiling water and then redrawn at 20° to the original drawing direction at 20° C. Arrows show the slips which are parallel to the *c* axis direction, [001] direction, of the first drawn structures. ×3.

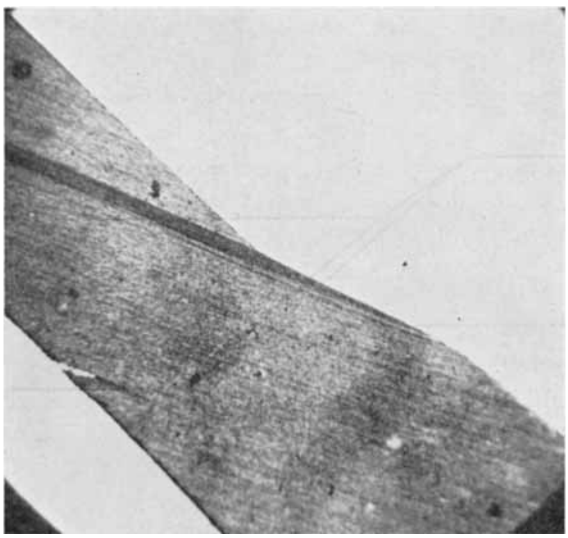


Fig. 4. Optical micrograph of a polyethylene film redrawn at 30° to the original drawn structure. ×30.

The difference of these two modes of deformation are shown schematically in Figure 2. Figure 2*a* shows the case where the film slips off. In this case it seems to be true that slips concentrate on one plane of original drawn structure and their directions are parallel to the [001] direction, that is, the *c*-axis direction of the original drawn structure. These slips are usually accompanied by a deformation band, the width of which increases with the increasing of the slipped length along the [001] direction of the original drawn structure. Also the [001] direction in this deformation band inclines a little to that of original drawn structure as described later. The angle of inclination, θ_1 in Figure 2*a*, which corresponds to the angle between the

[001] directions of drawn and redrawn structures, is about 6°, and the ratio of the slipped length L to the width of the deformation band D is constant and the following relation seems to exist.

$$D/L = \sin \theta_1$$

The values of θ_2, θ_3 are about 6° and 12°, respectively.

Figure 2*b* shows the case where plastic flow occurs. At the beginning the deformation is apparently similar to the uniform slip band observed on metals. But also in this case the [001] direction in the deformation band is parallel to the band and inclines at about 3–8° to that of original drawn structures. The angle φ_1 is about 6° and is equal to θ_2 and presumably to θ_1. As the elongation proceeds, this deformation band continues to be built up

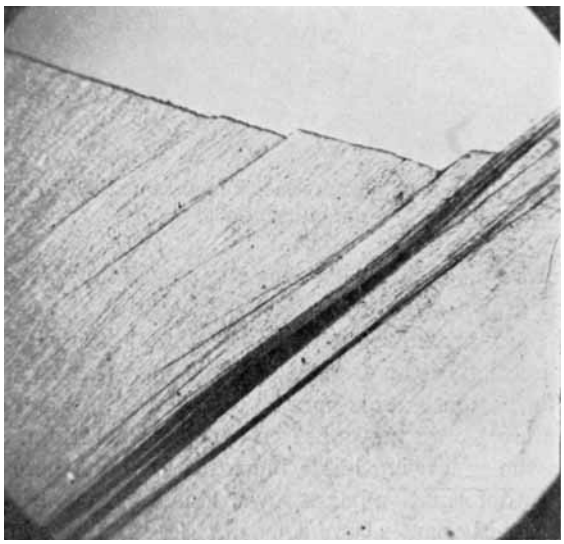


Fig. 5. Optical micrograph of a polyethylene film redrawn at 70° to the original drawn structure. ×60.

and lastly reaches a constant width, which forms the redrawn structure and remains constant during the plastic deformations. As shown in Figure 2*b*, the width of the redrawn region, D, depends upon the angles φ_1 and φ_2. The ratio of the width of redrawn structure to that of original drawn structure is given by the relation:

$$D/D_0 = \tan \varphi_1/\sin \varphi_2$$

where D_0 is the width of the original drawn structure, and φ_2 is equal to the angle between the original drawn and redrawn directions, from which the angle of inclination of [001] direction in the deformation band to the original structure is subtracted. There are given in Table I the D_0/D for various angle of redrawing. It is assumed for calculation that $\varphi_1 = 5°$ and the angle of inclination of [001] in deformation band is 5°.

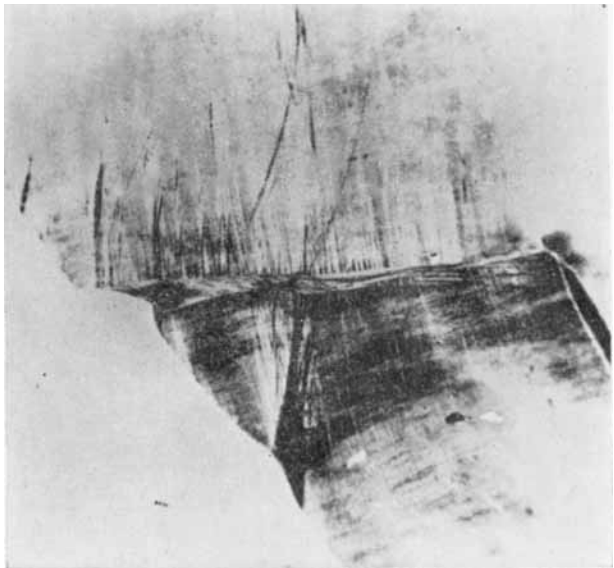


Fig. 6. Polarizing micrograph of polyethylene film redrawn at 80° to the original drawn structure. ×35.

Figure 3 shows a piece of film showing slipping which was redrawn at 20° to the original drawn structure. It was ascertained by microscopic observation and the x-ray diffraction that the direction of slipping is parallel to the [001] direction of the original drawn structure and the [001] direction in the deformation band is parallel to the band. When the redrawing angle becomes larger, we may observe a typical deformation band as in Figure 4. As the redrawing angle increases further, there may be seen groups of narrow deformation bands several microns in width at first, which are not always strictly parallel and some of them terminate in the film (Fig. 5). As the elongation proceeds, gradually the deformation band builds up and plastic flow begins to occur.

By a detailed microscopic examination, especially with a polarizing microscope it can readily be seen that slipped regions and their unslipped neighbors have somewhat different orientations. In a polarizing microscope with crossed Nicols, extinction positions are slightly rotated between slipped and unslipped regions, 3–8° in our experiments. The values coincide with the results of microscopic observation and the x-ray diffraction measurements.

As the slips shown in Figure 2*a* are usually accompanied by a deformation band which has a little inclined [001] direction, it will not be a simple slip. It is probably a complicated combination of slip and twin deformation. In the case of the homogeneous deformation band shown in Figure 2*b*, slips seem to occur on successive planes parallel to their [001] directions, but in the case shown in Figure 2*a*, slips seem to concentrate on one particular plane of original drawn structures. The result that the angles θ_1 and φ_1 are

TABLE I
Values of D_0/D[a]

Redrawing angle	φ_2	D_0/D Calculated	D_0/D Observed
20°	15°	3.0	—
30°	25°	4.9	5.0
40°	35°	6.6	7.2
50°	45°	8.1	8.0

[a] Samples used originally drawn 400% in boiling water.

nearly equal shows the fact that the mechanisms of both type of deformations are substantially the same.

When the angle of redrawing approaches a right angle to the original direction of drawing, for example 80°, simple slip deformations are no longer observed. In these cases many kink bands are formed nearly perpendicular to the original drawn structures, and then the structure with kink bands begins to flow through the relatively clear boundary and changes its orientation gradually to the direction of redrawing. Once flow has started, the stress decreases somewhat and remains nearly constant until the flow ceases. These kink bands are long and large enough to be observed with optical and polarizing microscopes and so seem to have no relation to the fibrillar or lamellar structures generally considered to be the constituents of the drawn structure. When the drawn films are redrawn at right angles to the original direction of drawing, the films redrawn at temperatures above 60°C. show the structure with kink bands clearly, though the films redrawn at a low temperature are easy to break off.

It is known that kink bands are formed when the original structures are compressed parallel to the direction of easy slip, so it may be taken that the increase of tensile component of load which occurs with the increase of redrawing angle to the original drawing direction gives rise to compressional force in the original drawn structure parallel to the [001] direction, the direction of easy slip, and forms the kink bands.

The details of the deformation mechanism of the formation of kink bands and successive flow have not been made clear, but perhaps it may be complicated deformation processes in which not only the (110) [001] slip system but also other possible slip systems and twin deformations join together. These deformations would not occur at random but have at least some regularity locally. Figure 7 is an electron micrograph of the necked region near the undrawn region of the films redrawn at right angles to the direction of original drawing, and shows a deformed kink band structure which is like a pointed twill figure.

The x-ray diffraction pattern of this position shows that there exist two fiber structures at angles to each other; the *c*-axis orientation in each of the two fiber structures is good, as estimated from the arc length of the diffraction patterns, and the angle of inclination between the two fiber struc-

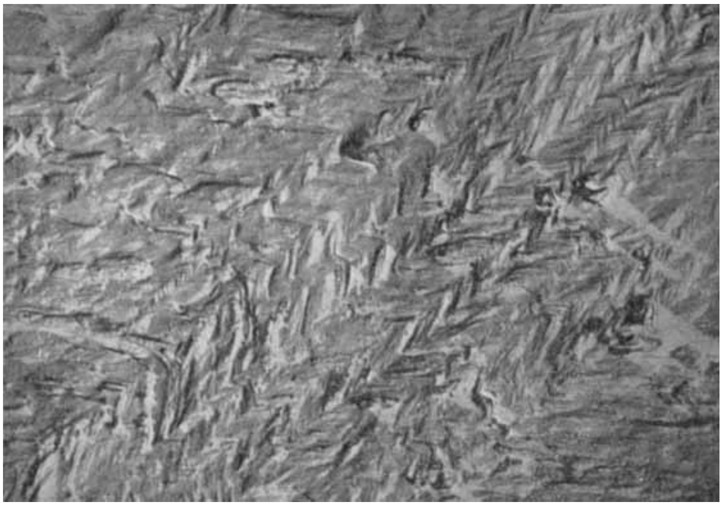
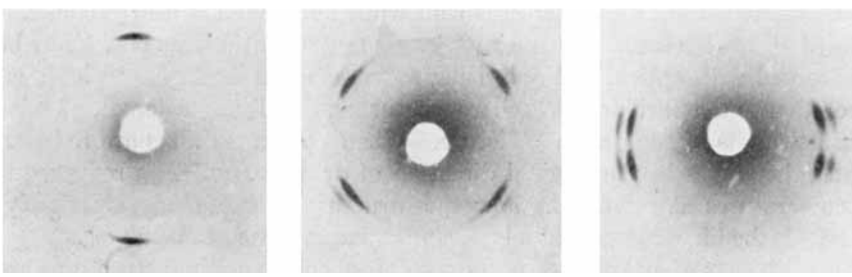


Fig. 7. Electron micrograph of a polyethylene film redrawn at right angles to the original drawn structure at 80°C. Direction of Cr-shadowing is parallel to the original drawing. ×7000.



(a) (b) (c)

Fig. 8. X-ray diffraction patterns of the same film as in Fig. 7; (a) original drawn structure; (b) upper part of neck, corresponding to Fig. 7; (c) lower part of neck. X-ray is vertical to the redrawing direction.

tures gradually decreases as the position illuminated by x-ray shifts to the more fully drawn region. The result that the intensities of the (200) patterns of redrawn regions increase in comparison with that of unredrawn portion of the film proves the existence of rotation which brings the (100) plane more normal to the plane of film. This rotation may be caused by the (100) [010] slip and (310) [$1\bar{3}0$] twinning.[1]

Figure 9 shows an electron micrograph near the boundary of slip bands in the films redrawn at 45° to the original direction of drawing. The slipped regions seem to be composed of more sharply and uniformly oriented fibrillar structures than that of unslipped regions but it is not clear whether these fibrillar structures represent the unit width of successive slip planes or not. The structure of the slipped region is inclined somewhat to that of unslipped region and there is no distinguishable change at the boundary.

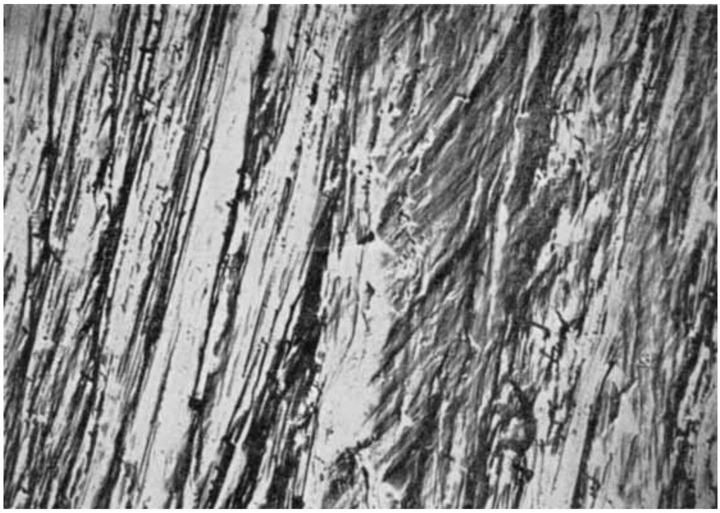


Fig. 9. Electron micrograph of the boundary of slip band in a film redrawn at 45° to the original structure which is drawn ×14 in boiling water. The right side of the micrograph shows the original drawn structure and the left the slipped regions. ×7000.

Figure 10 is an electron micrograph of the necked portion on the same film as Figure 6. The structure with kink bands changes to the redrawn structure through the edge nearly parallel to the original drawn structure, and the redrawn structure does not show a well oriented fibrillike appearance as that of the slipped regions in Figure 9. Also we can not see the pointed twilllike figure shown in Figure 7 because the elongation occurs selectively to one side owing to the fact that the redrawing direction inclines from the beginning.

The x-ray diffraction patterns of the redrawn region, corresponding to Figure 9, also show some changes, that is, the intensities of the (200) patterns increase a little and the equatorial patterns, corresponding to d = 4.5 A., appear as reported before[4] and disappear by annealing. The x-ray patterns corresponding to Figure 10 are of the two fiber diagrams shown as Figure 8, although one of them is very weak in this case.

From the above observations of the redrawing process of drawn polyethylene films it seems to be possible to conclude that slip and twin deformations take an important role in the plastic deformations in crystalline polymers just as in single crystals of metals, especially when they are well oriented and crystallized. Therefore one would be obliged to discard the well known fringed-micelle model and to conceive of a structure of the crystal having many defects.

Stress–Strain Behavior

The result of observations that slips and twins cause the plastic deformations in drawn polyethylene and their modes depend upon the direction of redrawing to the original drawing direction can be confirmed by stress–

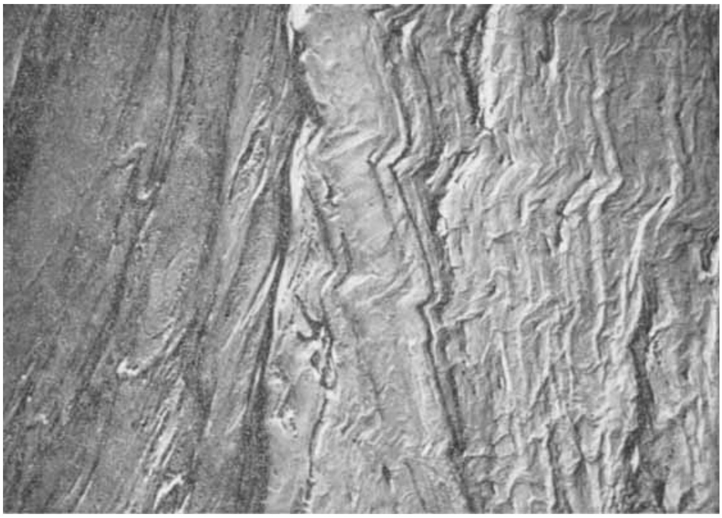


Fig. 10. Electron micrograph of the necked part in the same specimen as in Fig. 6. The right side of the micrograph shows the first drawn structure with kink bands, and the left side shows the redrawn structure. ×7000.

strain behavior of these films. Tables II and III show the value obtained from the stress–strain curves for two films drawn at different temperatures. Although these values depend upon extension rate, temperature, conditions of edges of the films and so on, one may obtain the general concept of the dependence of deformations upon the redrawing direction.

The films originally drawn in boiling water are slipped out when the angle between two successive drawing directions is small. If we assume that the slip plane is vertical to the film plane and the slip directions are [001]

TABLE II
Stress–Strain Behavior of Polyethylene Film Originally Drawn 1400% in Boiling Water (65% R.H., 20°C., rate of elongation 50%/min.)

Redrawing angle	Upper yield, kg./mm.^{2}	Lower yield, kg./mm.^{2}	Breaking strength, kg./mm.^{2}	Breaking extension, %	Type of extension	$f_{cr.}$ (calc.), kg./mm.^{2}
90°	—	—	2.9	—	Break off	—
80°	—	—	3.2	—	Break off	—
70°	—	—	3.5	—	Break off	—
60°	3.1	1.8	23.2	850	Slip band–flow–break	1.3
50°	3.5	2.4	31.1	910	Slip band–flow–break	1.6
40°	3.9	2.8	44.6	970	Slip band–flow–break	1.8
30°	5.7	—	—	—	Slip off	2.3
20°	7.2	—	—	—	Slip off	2.2
10°	15.1	—	—	—	Slip off	2.3

direction to the original drawn structures, the critical shear stress is approximately given by:

$$f_{cr} = (F/A)^1/_2 \sin 2\theta$$

where A is cross-sectional area, θ is the angle between two successive drawing directions, and F is the load applied. Calculated values of f_{cr} are also given in Tables II and III. When the angle between the two successive drawing direction is small and the films slip off, f_{cr} is nearly constant. However, we cannot decide what kind of deformation mode corresponds to this critical shear stress.

TABLE III

Stress–Strain Behavior of Polyethylene Film Originally Drawn 1000% at 30°C. (65% R.H., 20°C., rate of elongation 50%/min.)

Redrawing angle	Upper yield, kg./mm.^{2}	Lower yield, kg./mm.^{2}	Breaking strength, kg./mm.^{2}	Breaking extension, %	f_{cr}. (calc.), kg./mm.^{2}
90°	2.1	0.8	18.7	1400	–
80°	1.7	0.8	29.1	1450	0.27
70°	1.5	0.8	24.0	1400	0.45
60°	1.3	0.85	32.0	1350	0.54
50°	1.7	0.9	27.2	1300	0.50
40°	1.3	1.2	38.0	1200	0.63
30°	1.7	1.6	36.2	1760	0.72
20°	3.0	2.8	22.0	340	0.94
10°	7.3	7.1	15.7	40	1.17

The films redrawn at right angles break out by the time the plastic deformation begins. However, the temperature of elongation is higher, the film can be extended through the necked region, and one may observe clearly the kink band structures and pointed twill-like pattern on microscopic observation; the stress strain curves corresponding to this case shows the upper and lower yield remarkably.

When we redraw film originally drawn at low temperature, we cannot observe the slip or kink band visually or with a light microscope, but in the electron micrograph there can be seen the pointed twill-like figure, although locally and less clearly. The stress–strain curve of the film redrawn at right angles to the original direction of drawing shows a clear upper and lower yield. As the redrawing angle decreases, the necked region becomes less distinct, and at the same time the upper yield stress gradually decreases, although the lower yield stress remains constant. On redrawing at about 50°, the difference is smallest, and further decreasing of the redrawing angle cause the increase of both upper and lower yield stresses again. This stress–strain behavior seems to show the fact that the deformation mechanism of these films also varies with the redrawing direction and that there is no essential difference of the mechanism of drawing between the films drawn at high temperature and room temperature.

CONCLUSION

Observations with a light and an electron microscope on redrawing phenomena in drawn polyethylene films clearly show that their deformations are due to slips and twins similar to the case of metal and that the structure of well oriented and crystallized films are supposed to be similar to a single crystal. The mode of deformation depends upon the direction along which the stress of redrawing is applied. When the angle between first and second drawing is small, the (110) [001] slip system may occur, but as the redrawing angle approaches a right angle, other slips and twins take part in a deformation.

When the drawn films are redrawn at intermediate angles and extended with plastic flow, the ratio of the width of original drawn regions to redrawn regions is nearly constant and depends upon the angle between the upper side of the uniform deformation band to the direction of the band φ_1 in Figure 2*b*.

Stress-strain behavior of these films also supports the above discussion. When a film is redrawn at 50–90° to the original drawing direction, the lower yield stress is nearly constant, although the upper yield stress increases as the redrawing angle increases. It would be possible to think that in this range the deformation mechanism during plastic flow is much the same, and that the upper yield stress which depends upon the redrawing direction is necessary to make the direction of stress to incline at about 40° to the original drawn structure by forming the neck or kink bands.

The precise crystallographic meaning of this angle we can not decide now, but it would be probable that it has some relation to the result[5] reported that there is a tendency for the chain axis to be inclined to the fiber direction at a definite angle at first, when the polyethylene sample is drawn at room temperature.

If the shear stress which is necessary to begin a slip or a twin is larger than the breaking stress, a film breaks off by the time plastic deformation begins. But when the stress decreases to a value smaller than the breaking stress by means of inclining the redrawing direction or raising the redrawing temperature, plastic deformation can occur.

We wish to thank Mr. Sugimoto and Mr. Koosaka for their assistance in measurements.

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Résumé

Lorsqu'on relache le film de polyéthylène de haute densité complètement étiré, on peut observer une déformation de glissement, dépendant de l'angle de relâchement par rapport à la structure étirée originale. Lorsque l'angle de relâchement est très petit, il y a habituellement combinaison entre les glissements simples, une est la direction de la structure étirée originale (001), autre les bandes homogènes de glissement. Les directions (001) des régions qui ont glissé et celles qui n'ont pas glissé sont légèrement inclinées l'une vers l'autre. Lorsque l'angle de relâchement approche de 90°, on observe des bandes correspondants à des noeuds. De ces résultats nous pouvons conclure que la structure de ces films de polyéthylène étiré est très semblable à celle de cristaux simples avec défauts.

Zusammenfassung

Bei weiterer Reckung völlig gereckter Polyäthylenfolien hoher Dichte konnte eine Gleitungsdeformierung, die vom Winkel zwischen Zweitreckung und ursprünglicher Reckrichtung abhängt, beobachtet werden. Bei sehr kleinem Zweitreckungswinkel tritt gewöhnlich eine Kombination von Einzelgleitungen mit einer Richtung in (001) der ursprünglichen gereckten Struktur und einem homogenen Gleitungsband auf. Die (001)-Richtung von Gleitungs- und Nicht-Gleitungsbereichen sind gegeneinander schwach geneigt. Bei Annäherung des Zweitreckungswinkels in 90° werden Schleifenbänder beobachtet. Aus den Ergebnissen kann geschlossen werden, dass die Struktur dieser gereckten Polyäthylenfilme derjenigen eines Einkristalls mit Fehlstellen sehr ähnlich ist.

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