

Plastic Deformation of Polyethylene Terephthalate Films during Rolling

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Abstract—Amorphous polyethylene terephthalate was oriented via rolling on a laboratory mill under room conditions. The influence of rolling on the polymer structure was investigated. The plastic deformation of the polymer was inhomogeneous at the initial stages of rolling. The deformation was localized in a system of almost perpendicular shear bands forming a zigzag. In polarized light, the zigzag area of plastic flow looked like a regular system of parallel optical bands and possessing approximately identical thicknesses. The distance between the shear bands was approximately equal to double the thickness of the film. The development of the zigzag after compression was attributed to the true effect of strain softening of the polymer. While moving to the opposite surface, the perpendicular band emerged in the existing band after the release of shear bands on the film surface.

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INTRODUCTION

Orientational stretching is an effective way to increase the strength and elastic modulus of a linear polymer [1–5]. For instance, high-strength fibers are produced via stretching by dozens of times at slightly above the glass-transition temperature. Another method of orientation is the rolling of polymers [6–26], during which the polymer film is stretched in a continuous manner between two rolls rotating in opposite directions.

Rolling inhibits the brittle fracturing of polystyrene [17, 18]. According to [27, 28], the plastic flow of polymers during stretching below the glass-transition temperature develops in shear bands with a lowered density of the material and accordingly increases the free volume. As a result, the true shear stress decreases and further deformation is facilitated. This phenomenon is called *true strain softening*. Over time, the density of the material in the shear bands gradually increases, and the yield point increases to the initial value [17, 18].

A technique that makes it possible to visualize and characterize the structural transformations accompanying the various kinds of polymer deformation (uniaxial tension and compression, in-plane tension and compression, rolling, etc.) was described in [29–

32]. This technique consists of applying a nanometer metallic coating to the surface of a polymer.

Subsequent heating of the polymer results in its shrinkage, and deformation of the cover makes it possible to obtain information about the mechanism of plastic deformation of the polymer.

With the use of this method, the shrinkage during subsequent heating of amorphous polycarbonate pre-rolled at room temperature was investigated [31]. The deformation of polycarbonate during rolling was found to proceed heterogeneously, and there were areas with different directions of polymer orientation. Areas where the polymer was oriented in the direction perpendicular to the rolling axis were observed in PET. A system of alternating optical bands with widths on the order of hundreds of micrometers was detected via polarizing microscopy [33]. The bands were parallel to each other, were directed normal to the axis of rolling, and stretched from one edge of the polymer film to the other. The appearance of such structures during rolling of amorphous PET was not explained.

The purpose of this study is to investigate the nature of the inhomogeneity of plastic flow of films of amorphous PET in the process of rolling.

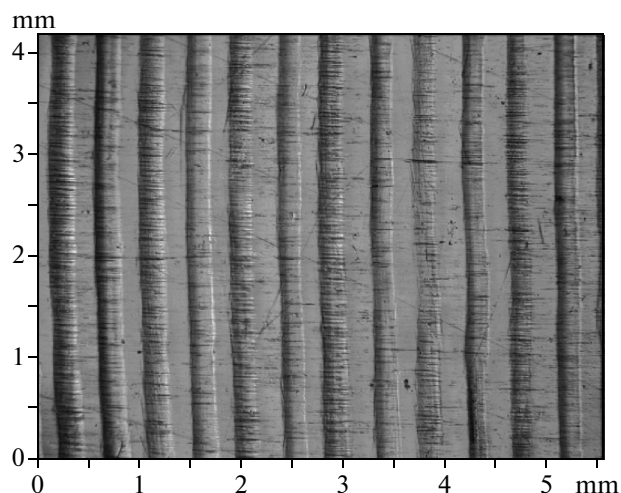


Fig. 1. A light micrograph of a sample of PET subjected to rolling up to 6%. The rolling direction is horizontal.

EXPERIMENTAL

Industrial isotropic amorphous films of PET with thicknesses of 190, 350, 510, and 910 μm were the objects of investigation. Film samples with a width of 18 and a length of 85 mm were subjected to rolling on a laboratory mill between two rotating rolls at room temperature to varying degrees of rolling from 1.02 to 1.42.

The degree of rolling, λ_{roll} , was evaluated as the ratio of the initial thickness, d_0 , to the thickness of the rolled sample, d . The thicknesses of the samples decreased in the studied interval of λ_{roll} owing to the extension of their lengths, while the widths remained almost the same.

Mechanical testing was performed on an Instron 4300 universal testing machine at a stretching rate of 5 mm/min. Samples in the form of bilateral blades with 6 mm $\times$ 20 mm working parts were cut with a knife along the direction of orientation.

Transparent samples of PET obtained via rolling were investigated under a light microscope (WOMO MBS-9, Soviet Union) and a Jeol JSM-5300LV scanning electron microscope.

Thin sections with thicknesses of ~ 10 μm were obtained from a rolled film of PET with the use of a microtome and photographed with a Floors R-112 polarizing microscope.

RESULTS AND DISCUSSION

Figure 1 presents a micrograph taken in transmitted polarized light of a PET film subjected to rolling up to 6% at room temperature. A system of regularly spaced optical bands formed as a result of the rolling of a transparent homogeneous film. The bands were parallel to each other, were directed normal to the axis of rolling, and stretched from one to the other end of the

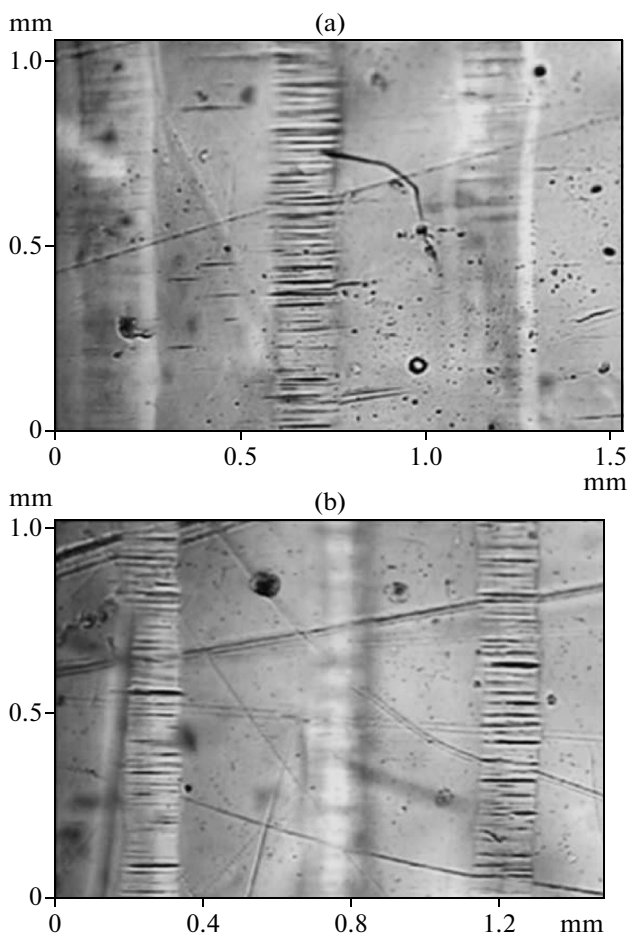


Fig. 2. The light micrograph of a fragment of the PET sample presented in Fig. 1 at a higher magnification: (a) focus on one side of the film, (b) focus on the back side of the film. The rolling direction is horizontal.

polymer film. The bands presented in Fig. 1 were located on different surfaces of the polymer film in a checkerboard pattern. Exactly half of the detected bands came to the focus when the microscope was focused on the surface of the film (every other one, Fig. 2a). Accordingly, the rest of the bands came to a focus when the microscope was focused on the second surface of the film (Fig. 2b).

The image of a thin slice of a laterally rolled PET film obtained via microtoming is presented in Fig. 3. A side cut was performed along the direction of rolling, which corresponded to the horizontal axis of Fig. 1. The image was inverted to remove the dark background. A sawtooth band running along the whole sample from the upper surface to the lower surface of the film and back was observed in the figure. Longitudinal bands appeared during microtoming of the sample with a knife. The sawtooth area was a region of intense plastic flow of the polymer during rolling. It shared triangular areas of plastically undeformed material. The shear bands oriented at an angle

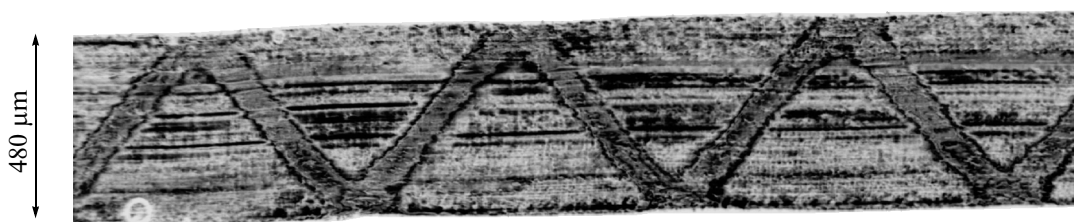


Fig. 3. Light micrograph of the surface of a PET film rolled up to 6% at room temperature.

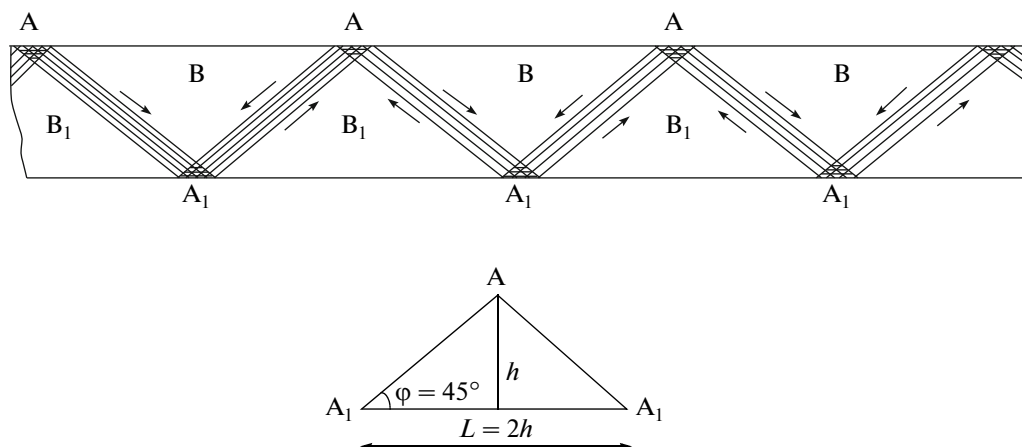


Fig. 4. Scheme of the mechanism of plastic deformation of the film during rolling. A and A₁ are the intersections of all bands; B and B₁ are undeformed areas.

$\varphi = 50^\circ \pm 1^\circ$ to the axis of rolling. This value was relatively close to 45° , which is typical for shear bands in polymers and metals. As a result, the adjacent yielded bands were almost perpendicular. The film surface after rolling was rough, and small holes were seen where the sawtooth band came out.

The mechanism of film deformation via rolling is schematically shown in Fig. 4. As a result of rolling, the film did not deform uniformly, but only in wide shear bands. The material in triangular areas B and B₁ between the shear bands after rolling did not deform. At the same time, the areas of intersection of shear bands, A and A₁, located on the opposite surfaces of the film, belonged to two bands and were severely deformed. This circumstance explains why areas A and A₁ in Fig. 1 in the transmitted polarized light looked different from areas B and B₁, where the light crossed only one of the shear bands.

The angle between the shear bands and the film plane was close to $\varphi = 45^\circ$ (Fig. 4), so the distance between the shear bands must have been equal to twice film thickness h :

$$L = 2h \quad (1)$$

The dependence of distance L between the shear bands disposed on one surface of the film on film thickness h is presented in Fig. 5. The points lie on a straight line going through almost the origin of coordinates. Thus, the distance between the shear bands was proportional to the film thickness. The slope of the straight line was 1.75 ± 0.05 , which is close to 2, the value predicted by Eq. (1). The difference of the slope of the straight line (1.75) from this value was due to angle φ being equal to 50° , not 45° .

The SEM-image of the surface of a PET film covered with a platinum coating with a thickness of 10 nm after rolling and heated to a temperature of 90°C , which was above the glass-transition temperature (75°C), is presented in Fig. 6. As a result of heating, shrinkage of the sample occurred, and its length decreased to the length before rolling. Because the platinum coating was very thin, it deformed together with the PET support and the identified areas of deformation during rolling. The platinum coating wrinkled owing to the shrinkage of the polymer support perpendicularly to the direction of shrinkage. According to Fig. 6, the film deformed nonuniformly during rolling, primarily in the light bands (shear bands). However, a small level of shrinkage of the polymer occurred between these bands, which were dark. A magnified image of the area between the

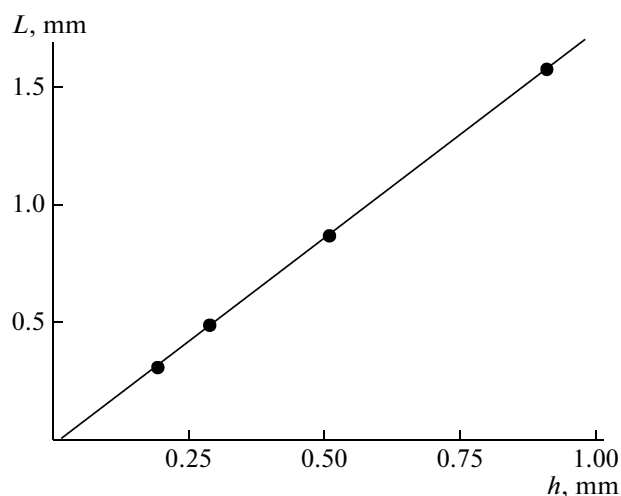


Fig. 5. Dependence of distance L between the shear bands disposed on one surface of the film on film thickness h .

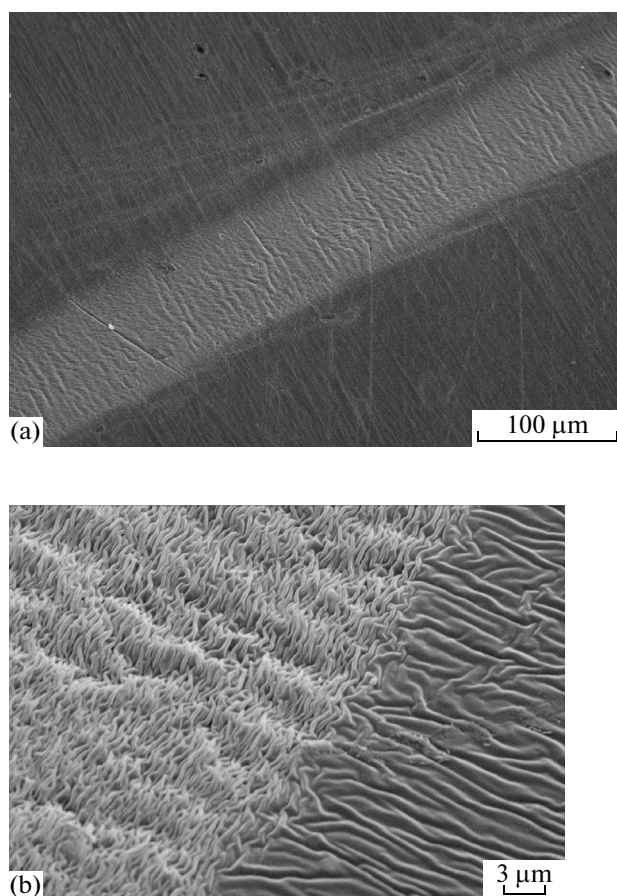


Fig. 6. (a) Scanning electron photograph of the surface of a PET film covered with a platinum coating with a thickness of 10 nm after rolling and then heated to a temperature of 90°C. (b) An enlarged view of the area on the band boundary.

bands is shown in Fig. 6b. Interestingly, the shrinkage in the dark bands appeared in the direction perpendicular to rolling.

CONCLUSIONS

New features of the morphology of amorphous PET that appeared during rolling of film samples have been discovered. The polymer deformed in mutually perpendicular shear bands that formed a zigzag. The size of the shear bands was on the order of the film thickness.

The appearance of a zigzag during compression in the process of rolling could be attributed to the softening of the polymer [17, 18]. A perpendicular band moving to the opposite surface emerged in the existing band after the release of shear bands on the film surface.

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