ORIENTATION PROFILE IN ROLLED POLYPROPYLENE

The application of such deformation processing techniques as rolling and drawing to polymeric materials is of great interest due to their ability to impart selective structure-property changes to the material at the same time dimensional changes are induced. In recent papers (1-4) the structure of rolled sheets of various polymers have been determined by such techniques as x-ray diffraction, microscopy, and light scattering, and the resulting structure correlated with various mechanical properties: anisotropic stress-strain response, impact strength, etc. Wilkes (5) has noted that wide-angle x-ray diffraction and low-angle light scattering patterns taken from near the surfaces of a rolled sheet differ markedly from those taken from near the center, and suggests a variation in morphology through the thickness direction of the sheet. Such a variation is obviously an important aspect of structure-property studies of rolling, but has received little attention in these prior studies. This letter discusses the deformation profile predicted by slip-line field plasticity analyses of rolling, and presents experimental confirmation of the results for rolled polypropylene.

The flow of material undergoing deformation in a rolling mill is governed by a set of hyperbolic partial differential equations obeying boundary conditions relative to both stresses and velocities. Although these equations can be solved for certain cases of plastic flow by means of a numerical method due to Hill (6), rolling is best analyzed by the trial-and-error approach developed by Prager (7). Here one assumes the shape of a portion of a slip-line field (a mapping of maximum shear stress directions) and then develops the remainder of the field so as to maintain compatibility between the associated stress field and the stress boundary conditions. The trial solution so obtained is then checked for compatibility with the velocity boundary conditions by constructing a hodogram (velocity field) from the slip-line field. Successive trial solutions are generated until compatibility is obtained. These solutions generally assume a homogeneous, isotropic, rigid-plastic material which does not exhibit strain-hardening or Bauschinger effect and whose response is independent of strain rate or temperature.

Alexander (8) has employed this geometrical method to develop a slip-line field for the hot-rolling process (where the temperature is such that strain hardening is nullified by thermal softening), assuming the absence of slippage between the material and the rolls over the arc of contact. Johnson and Kudo (9) later adapted this solution to determine the temperature profile in the roll gap and the distortion of a square grid. Certain of the results of these analyses may be summarized as follows: a) Material enters the roll gap at a velocity much less, and leaves at a velocity slightly higher, than the peripheral velocity of the rolls. These two regions are separated by a "neutral zone" of dead material which rotates as if part of the roll. b) A thin rigid slice of ma-

terial is predicted to exist at the entry point, the boundary of which would be expected to cause a band of intense shearing near the surface of the rolled sheet. c) In the entry zone, there is a progressive retardation of inner layers of the sheet relative to the outer layers, such that material deformation varies in an approximately parabolic manner from a minimum at the center of the sheet to a maximum at the outer surfaces. After leaving the neutral zone, the material experiences a slight reversal of this distortion.

A sheet subjected to multiple rolling could be expected to exhibit a deformation profile similar in form but of greater magnitude than the above solution: i.e., an approximately parabolic variation, symmetrical with the center of the sheet, and showing some evidence of discontinuity near the surfaces as a result of the shearing zones at the roll entry. In a polymeric material, where flow in general serves to produce molecular orientation, this result should be manifest in a similar variation in orientation through the thickness of the sheet.

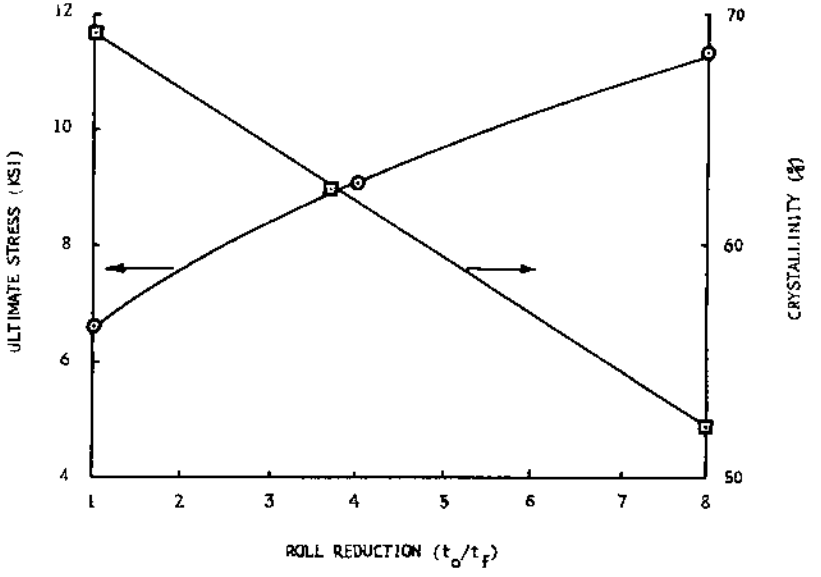

Fig. 1. Variation of crystallinity and strength with roll reduction.

As part of a larger study of deformation of polymers, we have subjected sheets of commercial molded polypropylene to multiple biaxial rolling (90° rotation of piece between passes) to various total thickness reductions of up to 8:1 using a variable speed Fenn mill having 12.5 in. diam rolls. The thickness reduction per pass was approximately 5%, and the temperature buildup due to deformation was limited to approximately 120°F by quenching the piece as necessary.

As reported earlier (10) biaxial rolling produces a uniplanar molecular orien-

tation (molecular chains lying perferentially in planes parallel to the rolling plane and randomly oriented in this plane). This structure is manifest in a tendancy of the rolled piece to delaminate, and in fact delamination limits the total thickness reduction possible to approximately 8:1. Rolling is an essentially traumatic process, and appears to reduce crystallinity in the piece by breaking up crystalline regions. Figure 1 shows the variation in crystallinity (as calculated from gradient-column density measurements) with rolling thickness reduction. Since a greater number of molecular chains are available as load-bearers, tensile pieces cut from the rolled sheet show increased strength; the variation of tensile strength with thickness reduction is also shown in Figure 1. Rolling reduces the fracture toughness for quasistatic loading, but at higher loading rates the delamination mechanism serves to increase the energy absorption of rolled material over unrolled: the ballistic impact resistance can be improved by nearly 30% by rolling to 8:1 thickness reduction.

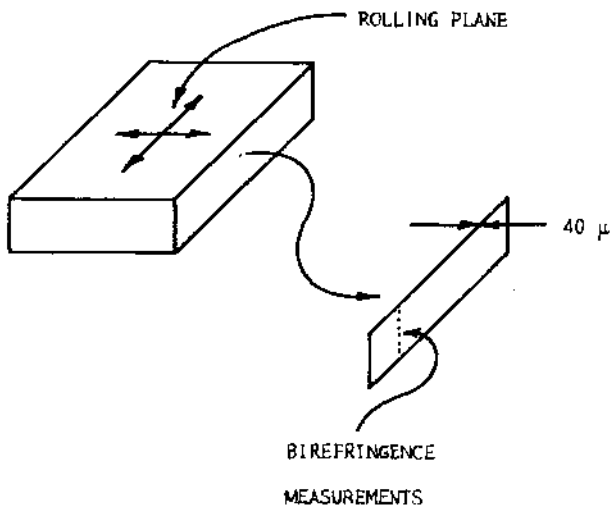

Fig. 2. Location of birefringence measurements.

The variation in molecular orientation through the thickness direction of the rolled sheet can be measured conveniently by means of birefringence microscopy: as illustrated in Figure 2, a thin (40 μ) ribbon is microtomed from the edge of a piece cut from the rolled sheet and the optical retardation is then measured at various points in the ribbon using a polarizing microscope in the Senarmont configuration. Although this birefringence is made up of crystalline, amorphous, and form contributions and a rigorous calculation of orientation functions would require independent data (e.g., sonic modulus or x-ray diffraction) at each small point in the ribbon, the birefringence itself is a fair measure of the total orientation of the sample point and will suffice for our present purposes.

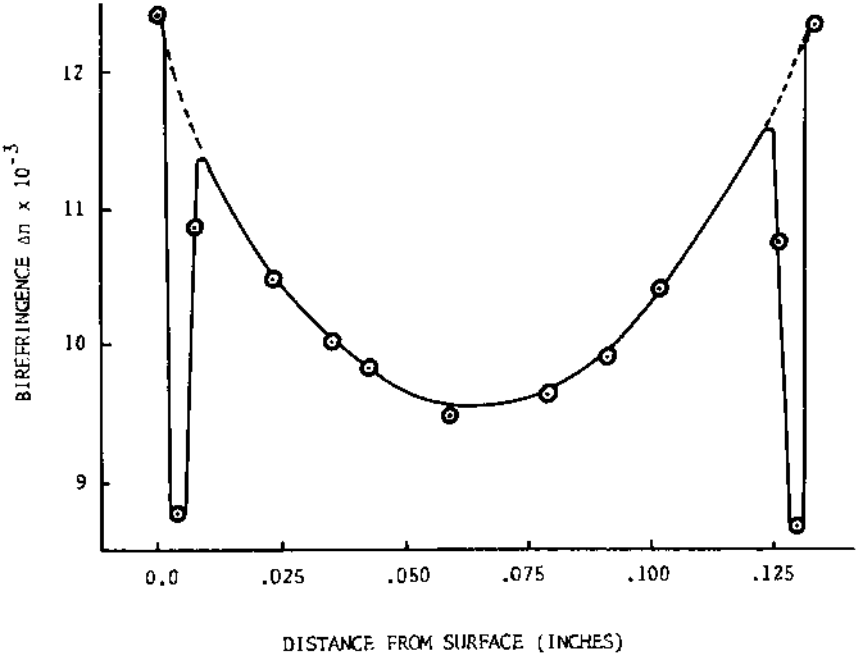

Fig. 3. Orientation profile through thickness direction in rolled polypropylene.

Figure 3 is a plot of measured birefringence as a function of depth in the thickness direction of the rolled sheet. An approximately parabolic variation in birefringence is obvious, with marked intensity minima superimposed near the edges. These minima appear as distinct bands in Senarmont micrographs, and are at approximately the location predicted for the bands of intense shearing in the Alexander soultion. The parabolic profile is in agreement with the deformation profile predicted analytically, and the bands of lesser birefringence can tentatively be ascribed to chain relaxation arising from localized heating or extensive bond rupture due to the shearing.

Similar results have been obtained for rolled nylon, and it is likely that most thermoplastics rolled above their T_g will exhibit a similar variation of orientation through the thickness direction. Previously reported orientation studies of rolled polymers should be interpreted with this in mind. Finally, it is worth mentioning that the measured birefringence profile of Figure 3 provides a confirmation of the Alexander solution which does not require alteration of the material's continuity before rolling, a drawback to the internal-inscribed-grid technique used in the past (11).

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