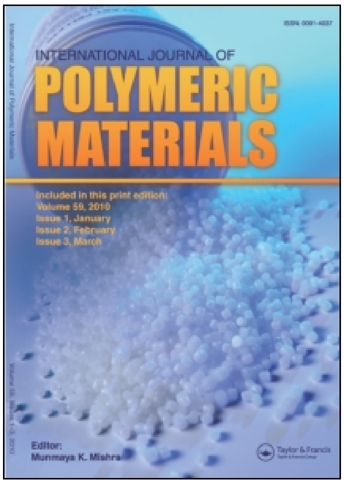

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The Properties of Pipes Produced under the Conditions of Spiral Flow

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The present communication is concerned with the relation between the strength properties and the morphology of low- and high-density polyethylenes in the extrusion of pipes under the conditions of spiral flow. It has been found that the optimal strength properties of pipes result when lamellar crystals are oriented in the middle portion of the pipe wall about 45° relative to its axis. Model experiments have been carried out on polystyrene melts through the use of optical-polarization technique and, as a result, the data obtained for polycrystalline polymers have been verified from the qualitative point of view.

INTRODUCTION

The investigations of many workers are directed to the search for methods of fabrication of pipes possessing high strength characteristics,[1–5] which are largely governed by the morphology of the polymer. Significant results in this line have been obtained by controlling the direction of flow of the melt, whereby the macromolecules are oriented so that the direction of the texture in the article coincides maximally with the isostaths of the stress field of loaded articles.[6–10] Wide possibilities of controlling the structure of polymer in pipes are provided in the extrusion of pipes in the spiral flow.[9–15] Indeed, by

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changing the relative speed of the axial and peripheral flow, that is, by changing the spiral pitch, it is easy to attain various degrees of orientation of macromolecules in the axial and peripheral directions, which must influence substantially the morphology of polymer in the article. The spiral flow in the extrusion process can easily be provided by specifying the rotation of the mandrel in the extruder head.[9,10,15–18]

The aim of the present work was to examine the relationship between the strength properties and the morphology of polymer as a function of the character of orientation assumed by the polymer melt in the spiral flow and partially fixed upon subsequent crystallization. Besides, experiments have been carried on an amorphous polymer, polystyrene, with a view to making use of the polarization optical method for controlling the process of orientation in the polymeric material while it is flowing in the screw.

EXPERIMENTAL

We have investigated low-density polyethylene with a molecular weight of 30,000 and a melt index of $i = 2.0$ g/10 min and high-density polyethylene with a molecular weight of 90,000 and a melt index of $i = 0.6$ g/10 min (in both cases the melt index was determined according to the ISO procedure).

Pipes of 25 mm diameter were fabricated in an extruder in whose head the mandrel was rotated by a direct-current motor through a reduction gear-box at a speed of 0 to 50 revolutions per minute.[14] The mandrel used had the following dimensions: the diameter of its cylindrical part was 25 mm, its length 85 mm, and the thickness of the moulding clearance 2.5 mm. The temperature of the melt in the moulding clearance was: 145°C for low-density polyethylene and 180°C for high-density polyethylene. The rotational speed of the extruder screw and the other parameters of the process were kept constant.

The polyethylene pipes produced were subjected to tensile tests. For this purpose, specimens were made from pipes, the design length of the working part being 25 mm; these specimens were cut out by means of a forming die in the longitudinal and lateral directions relative to the pipe axis. The speed of the moving grip of the tensile machine was specified at 100 mm per minute.

To measure the short-term strength in hydrostatic tension, pipe specimens having a length of 15 diameters were tested on a bench according to the "loaded-end" scheme. The test conditions were chosen so as to break the pipe in 10 to 15 seconds.[19]

The long-term strength (static fatigue) was determined on the bench according to the "nonloaded-end" scheme at a temperature of 20°C. The test pressure was calculated separately for each specimen according to the Nadai formula,[10] and the load coefficient was changed within the limits of 0.7 to 1.0.

The critical stress in the pipe wall, at which it was broken for 10 to 15 sec, was 250 kgf/cm^2 for pipes of low-density polyethylene and 570 kgf/cm^2 for pipes of high-density polyethylene.

Electron-microscope studies were carried out for various sections of the pipe specimen. In order to control the orientation of structural formations the direction of shading chosen was across the direction of extrusion for longitudinal sections and along the radius of the pipe for lateral sections. The surface layers of the objects under investigation were subjected to sublimation in the plasma of a gas discharge and then the underlying layers were etched in the gas discharge. Vacuum-sprayed replica was applied onto the etched surfaces by the well-known procedures.[21]

Wide-angle diffraction patterns of X-ray scattering were obtained with copper radiation using a Ni filter.

In addition to the electron-microscope investigations, the process of orientation of the extrudate was examined by the polarization-optical method as well.[22] The model material used in these experiments was polystyrene. In order to simulate the conditions of flow of polyethylene as closely as possible, polystyrene and its flow regime were chosen so that the apparent viscosities of the melt were approximately equal.[23] The extrudate in the form of tubular blanks forced out with the immovable and rotating mandrels was rapidly cooled and sections were prepared from it perpendicularly to and parallel with the generatrix. The specimens were ground to a thickness of 1.5 ± 0.003 mm and polished. The coefficient of optical sensitivity with respect to stress was determined by the procedure reported in the literature.[24] The ray path difference in the specimens was measured by the compensation method.[25]

RESULTS AND DISCUSSION

For pipes of high-density polyethylene (Figure 1) the yield value (curve 1) and tensile strength (curve 2) in the lateral direction increase with increasing number of revolutions of the mandrel, while those (curves 3 and 4, respectively) in the longitudinal direction decrease. The magnitude of relative elongation of pipe specimens (Figure 2) also increases in the lateral (curve 1) and decreases in the longitudinal direction (curve 2). The short-time strength of pipes in hydrostatic tension (Figure 2, curve 3) increases by 20 per cent, the optimum of strength properties being observed at a rotational speed of the mandrel $n = 20$–25 rpm.

The strength properties of pipes of low-density polyethylene fabricated with the mandrel being immovable and rotated varies in a manner (Figures 3 and 4) analogous to that described above, but the quantitative ratio of the strength characteristics is somewhat different.

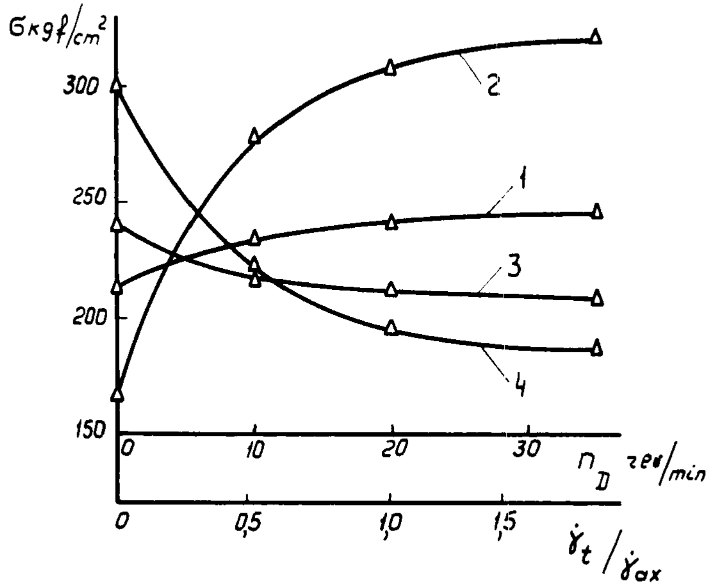


FIGURE 1 Dependence of the yield value and tensile strength of pipes of high-density polyethylene on the rotational speed of the mandrel.

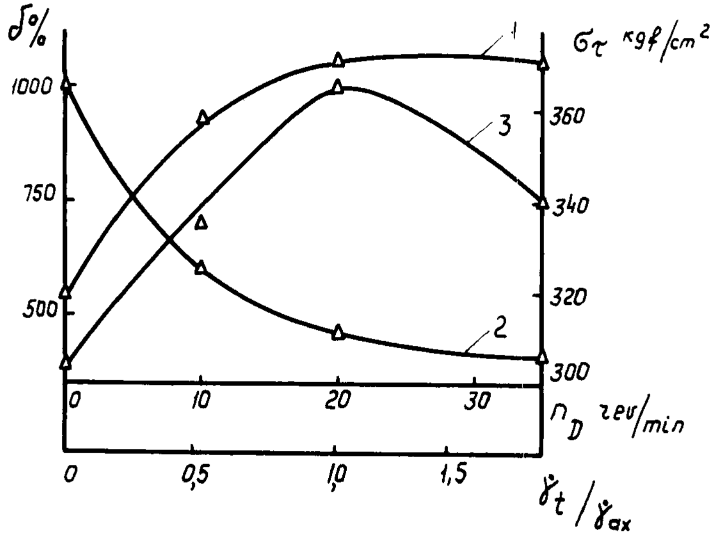


FIGURE 2 Dependence of the relative elongation in extension and the ultimate strength in hydrostatic tension of pipes of high-density polyethylene on the speed of rotation of the mandrel.

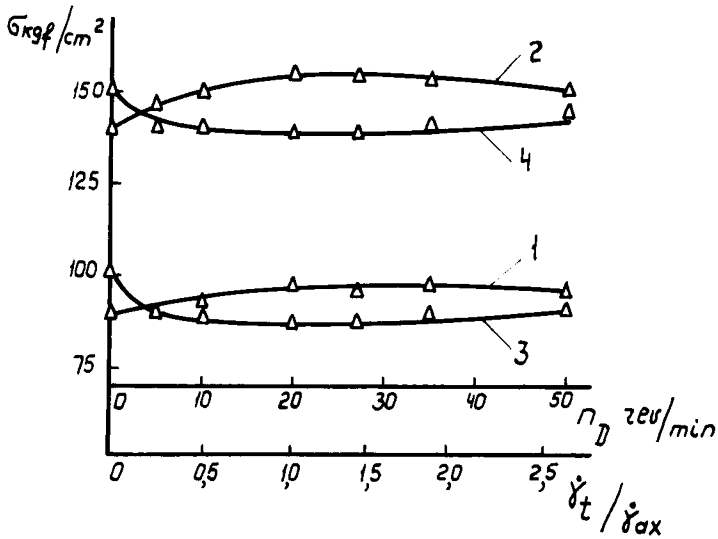


FIGURE 3 Dependence of the yield value and tensile strength of pipes of low-density polyethylene on the rotational speed of the mandrel.

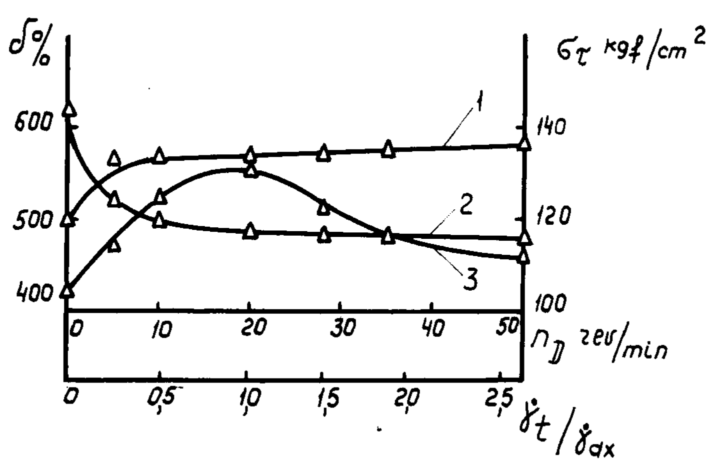


FIGURE 4 Dependence of the relative elongation in extension and the ultimate strength in hydrostatic tension of pipes of low-density polyethylene on the rotational speed of the mandrel.

B

It is interesting to note that the properties of pipes fabricated at the ratio of the rates of shear of the axial and peripheral flows being within the limits 0.15 to 0.2 exhibit isotropy. This is probably due to such conditions of flow of the melt into the extruder head in which there is attained a sort of two-axial orientation of the melt.

Another interesting feature in the properties of pipes is the increase of the tensile strength during the extension test of pipe specimens, which is accompanied by an increase in the relative elongation. This may be associated with the slipping of lamellar crystals and crystallites during the test.

From experimental data on the long-term strength of pipes of high-density polyethylene, which are fabricated at various rotational speeds of the mandrel it follows that the relation $\sigma = f(ln\ \tau)$, where σ is the stress in the pipe wall and τ is the time before rupture, is characterized by a monotonic change of the curve with a relatively abrupt fall at small values of durability. The nonlinear variation of $\sigma = f(ln\ \tau)$ must not cause any doubt as to the validity of the thermofluctuating nature of fracture,[26] since the observed deviation from the routine relation may be ascribed to the complexity of the loading conditions in which the specimens are loaded by the internal pressure, and also to the complex spatial arrangement in the polymer structure, which may have been the cause of the inconstancy of the structure coefficient γ in the equation defining the long-term strength of the material, suggested by Zhurkov.[27] It should be noted that there exists an opinion, according to which the structure coefficient depends, in a number of cases, on temperature and stress, and the insertion of the function $\gamma = \phi(\sigma, T)$ into the Zhurkov equation results in the smoothing out of the durability curve in the coordinates of σ versus $ln\ \tau$.

The tests carried out have shown that the best strength properties are displayed by pipes made at $n = 20$ rpm (curve 2 in Figure 5). With the stress

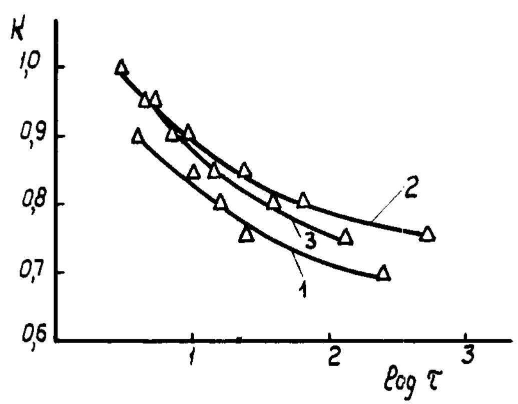
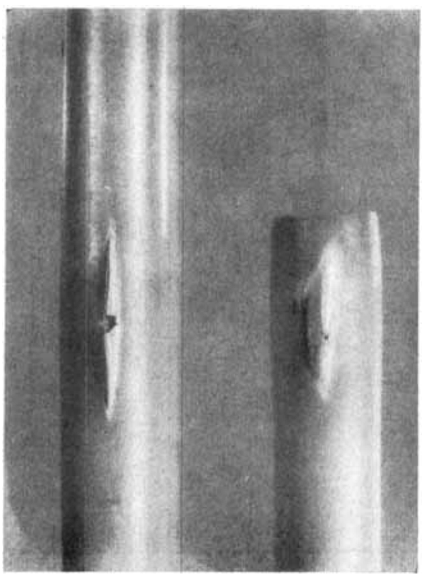


FIGURE 5 Durability curves for pipes of high-density polyethylene (1—$n = 0$; 2—$n = 20$ rpm; 3—$n = 35$ rpm).

in the pipe wall being equal, that is, with $\sigma = 427$ kg/cm², $ln\tau = 2.7$ for pipes produced at $n = 20$ rpm, and for pipes fabricated at $n = 0$ rpm, $ln\tau = 1.6$. This corresponds to the increase of the endurance time under loading before fracture of the pipe specimen from 40 min ($n = 0$) to 500 min ($n = 20$ rpm). As the load increases the curves diverge and the difference in the time, on the basis of which the load-bearing capacity of the pipe is estimated, increases. A further increase in the number of revolutions of the mandrel (say, $n = 35$ rpm) however leads to a decrease in the durability of pipes (curve 3). This is especially manifested when the mandrel rotates at 40 rpm.

An interesting feature of pipes fabricated with the mandrel being rotated is the change in the direction of the fracture line at breakdown of the specimen relative to its axis. For ordinary pipes fabricated with the mandrel being immovable the fracture line formed is parallel to its axis (Figure 6a), and for pipes fabricated with the mandrel being rotated the fracture develops at an angle α to the pipe axis (Figure 6b). In this case, with increasing rotational speed of the mandrel the angle of deviation α sharply increases and then remains practically unchanged (Figure 7).

The curves of durability and the change of the fracture angle for pipes of low-density polyethylene (Figures 8 and 9) do not differ qualitatively from those for pipes of high-density polyethylene.



a b

FIGURE 6 Photograph of specimens of pipes of high-density polyethylene after durability test (a—mandrel being immovable; b—mandrel being rotated).

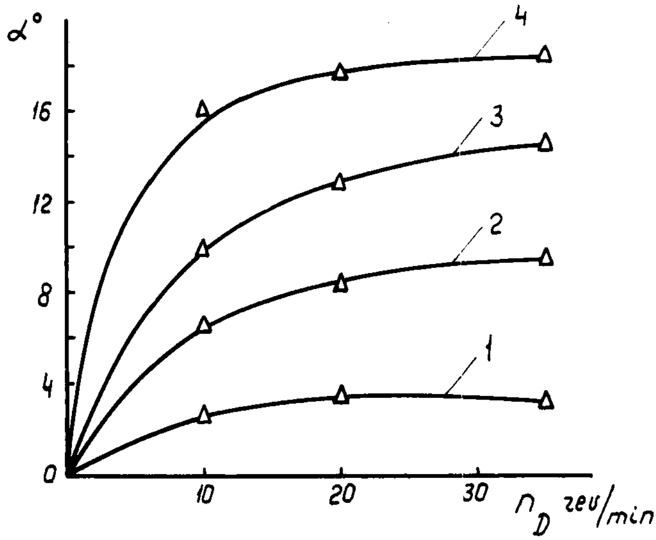


FIGURE 7 Dependence of the angle of deviation of the fracture line on the rotational speed of the mandrel for pipes of high-density polyethylene (1—K = 1.0; 2—K = 0.9; 3—K = 0.85; 4—K = 0.8).

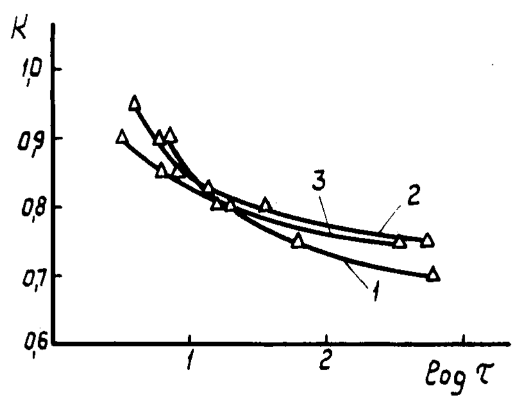


FIGURE 8 Durability curves for pipes of low-density polyethylene (1—n = 0; 2—n = 20 rpm; 3—n = 35 rpm).

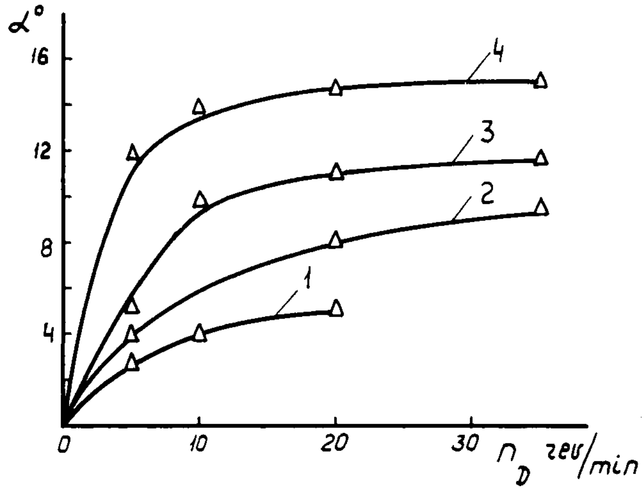


FIGURE 9 Dependence of the angle of deviation of the fracture line on the rotational speed of the mandrel for pipes of low-density polyethylene (1—$K = 0.95$; 2—$K = 0.90$; 3—$K = 0.85$; 4—$K = 0.75$).

Pipes made by conventional methods are uniaxially oriented and they are characterized by an axial texture. This results in such an anisotropy of properties, at which pipes possess the best strength characteristics in the axial direction. The tendency in practice is to reduce this anisotropy to a minimum and thereby to balance the properties in different directions. The data given above show that the strength characteristics of pipes can be considerably improved if pipes are fabricated under the conditions of spiral flow. The optimum strength of pipes is attained if the ratio of the rates of the axial and peripheral flows is 1:1.

Interesting results were obtained in electron-microscopic investigations. No spherulite structure was detected in the specimens under study. On the other hand, the canonical types of morphology detected were large layered formations (macrolayers), the fine structure of which may also be presented as consisting of layered formations—lamellar crystals. According to the Hosemann model, the arrangement of lamellar layers in an oriented material must be orthogonal to the direction of orientation of polymers.[28] Thus, it might be expected that the crystalline structure would indicate the local directions of the force fields in any section of the pipe. Therefore, in an analysis of photographs the direction perpendicular to the layer width (to the plane 001 of a lamellar crystal) was assumed to be the local direction of the force field.

The microphotographs of the pipes produced at $n = 0$ (Figure 10a and b) show that the crude morphology of polymer may be presented as consisting of

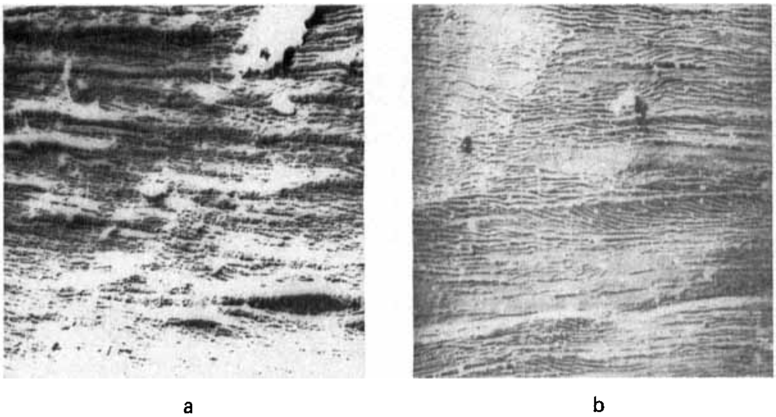


FIGURE 10 Microphotographs of a pipe fabricated with the mandrel being fixed (a—cross section; b—longitudinal section).

layered formations both in the longitudinal and in the lateral direction. In the latter case they are in the form of concentric rings resembling annual growth rings in the cross-section of wood. In the longitudinal section the layered character of morphology is analogous to the high-oriented state of uniaxially oriented polymers. There is, however, a difference in the processes of structure formation of oriented polymers in extension, for example, the processes involved in the formation of fibres[29] are different from those taking place in the extrusion of pipes considered in the present work.

If pipes are fabricated by rotation of the mandrel, the specified orientation of the polymer melt is such that upon subsequent crystallization the lamellar crystals and even the macrolayers are locally rearranged so that the direction of their thickness coincides with the direction of the vectors of the rates of shear of axial and peripheral flows (Figure 11a and b). According to our data (for $n = 20$ rpm) the angular position of lamellar crystals in the middle of the pipe wall is estimated at 45° relative to the extrusion direction. Diffraction patterns of X-ray scattering for pipe specimens near the inner and outer surfaces (Figure 12a, b, c, and d) points to the presence of a noticeable texture. It is characteristic that for pipes produced at a rotational speed of the mandrel of 20 rpm, the direction of texture at the inner surface of the wall (Figure 12d) changes by about 90°.

The results obtained from model tests on polystyrene are as follows. In a section perpendicular to the generatrix of a tubular blank fabricated with the mandrel being immovable, the path difference has a maximal value in the middle of the wall and decreases near the inner and outer surfaces by a para-

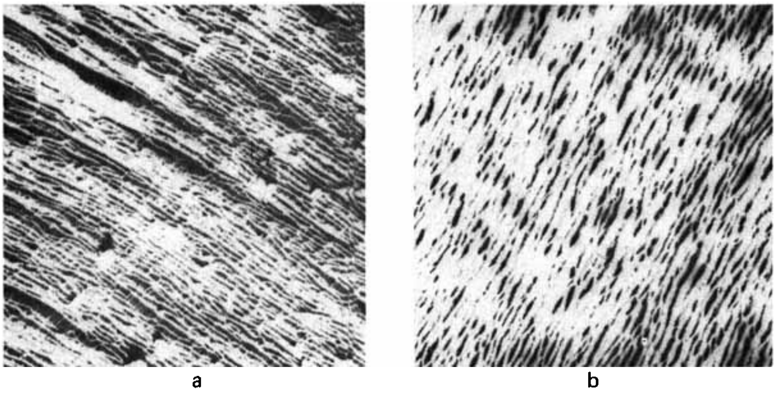


FIGURE 11 Microphotographs of a pipe fabricated with the rotational speed of the mandrel being 20 rpm (a—cross section; b—longitudinal section).

bolic law (Figure 13a). In a section parallel to the generatrix the material was found to be uniaxially oriented since it is characterized by the zero parameter of the isocline. This is verified by the special case of the Mesnager theorem.[30] The principal stresses in the inner and outer surfaces of the wall of a tubular blank, which are in contact with the working parts of the extruder head, are negative in sign (compressive stresses along the orientation), and in the middle of the wall they are positive (tensile stresses); the latter are lower in magnitude than the compressive stresses (Figure 13b). Thus, a large degree of orientation of macromolecules in the direction of the axis of the tubular blank along the edges of the wall and their partial disorientation in the middle part are provided by the conditions of movement of the melt in the axial flow.

The path difference acquired by the components of the polarized light in passing through a section that is perpendicular to the generatrix of the tubular blank made at $n = 10$ rpm (for the given extrusion conditions the ratio of the rates of shear of the axial and peripheral flows is 1:1) becomes maximal on the inner surface of the pipe and is the lowest on the outer surface (Figure 14a). In a section parallel to the generatrix of this blank the picture of quasi-principal stresses is also changed (Figure 14b). From these data it follows that near the outer surface of the wall the direction of the major axis of the ellipsoid of the macromolecule practically coincides with the axis of the tubular blank, and near the inner surface it develops perpendicularly to it. Analogous results are presented in Figure 14c and d.

A comparison of the information concerning the flow of the melt in the extruder head and of the data obtained by the polarization-optical, electron-

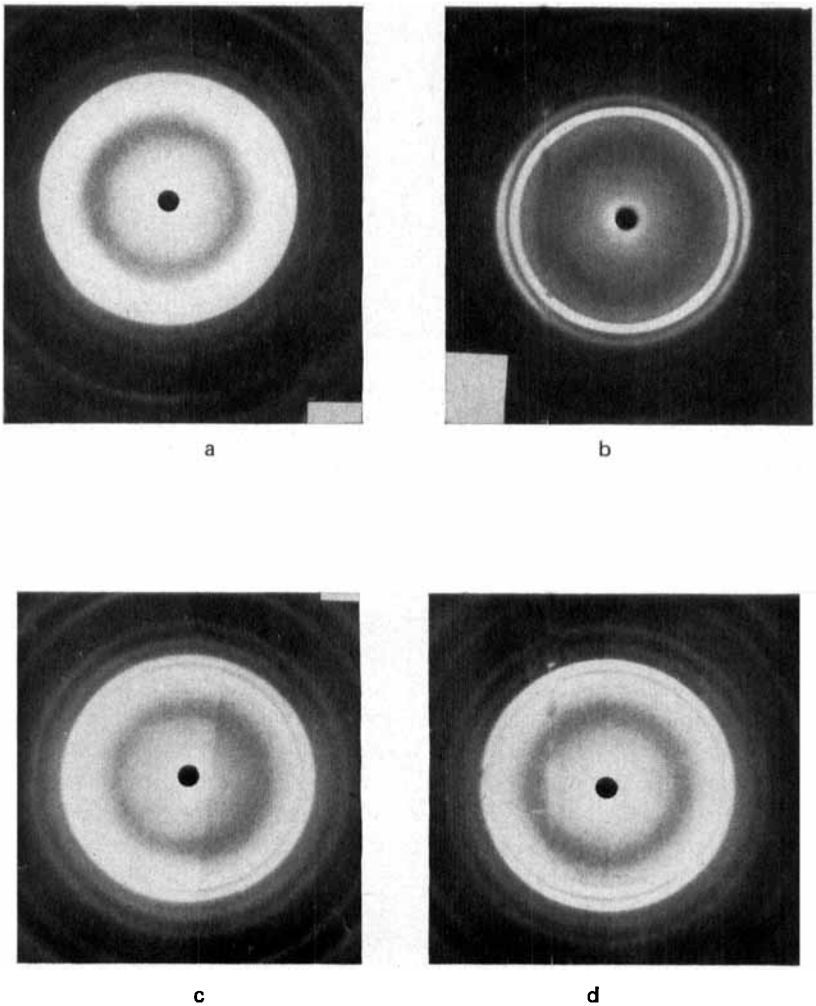


FIGURE 12 X-ray patterns of pipes of high-density polyethylene: a—$n = 0$, outer layer; b—$n = 0$, inner layer; c—$n = 20$ rpm, outer layer; d—$n = 20$ rpm, inner layer.

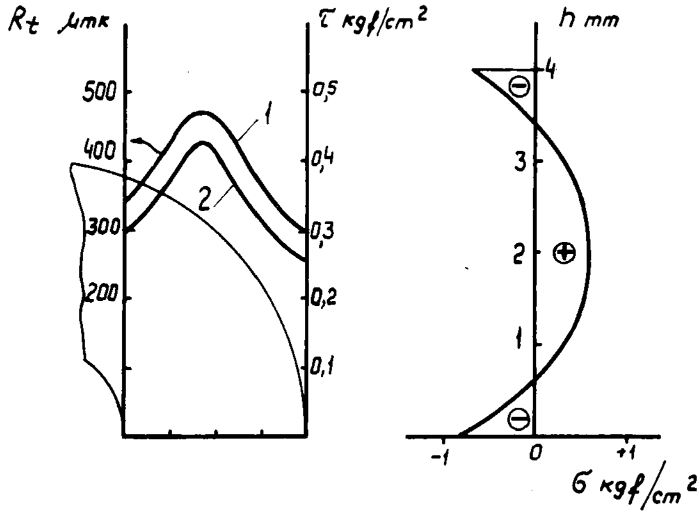


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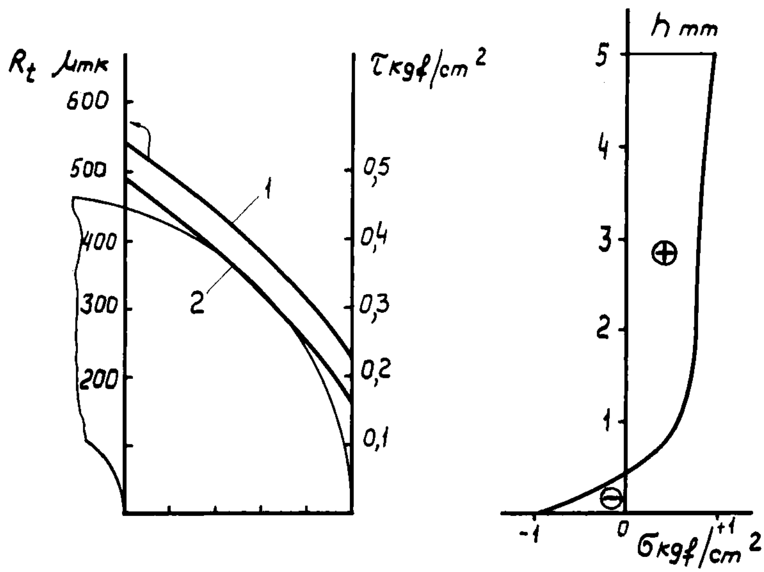
FIGURE 13 Diagrams of the path difference and of the tangential and principal stresses in a tubular blank made with the mandrel being immovable: a—cross section; b—longitudinal section.



a b

FIGURE 14 Diagrams of the path difference and of the tangential and quasi-principal stresses in a tubular blank fabricated with the rotational speed of the mandrel being 10 rpm: a—cross section; b—longitudinal section.

microscopic, X-ray and mechanical methods of investigation shows that they are consistent, enabling one to obtain a sufficiently full picture of the phenomena observed in the extrusion of articles through the use of spiral flow.

CONCLUSION

The optimum strength properties of pipes are provided if they are fabricated at a ratio of the rates of shear of the axial and peripheral flows of 1 : 1. An increase in this ratio leads to a decrease in the strength of pipes being fabricated. The spiral flow of the melt creates conditions under which the lamellar crystals and macrolayers of the pipe material are found to be oriented in the direction of the vectors of the rates of shear of the spiral flow. In this flow the macromolecules near the outer surface of the wall of a pipe are oriented along the generatrix, and near the inner surface they tend to be oriented perpendicularly to it. Thus, the use of the mandrel rotation in the extruder head makes it possible to alter the service properties of pipes by controlling the morphology of polymer without substantially changing the technological process.

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