F. Ania, R. K. Bayer*, F. J. Baltà Calleja

Instituto de Estructura de la Materia, Madrid, Spain

Biaxially-oriented Polyethylene Films by Compression of Injected Moldings

Biaxially oriented PE films were prepared by compression of uniaxially oriented elongational-flow injection-molded precursors. The films so prepared, having different extents of biaxial stretching were compared on the basis of two derived functions, biaxial draw ratio ($\bar{\lambda}$) and degree of biaxiality (B). The former gives a quantitative measure of the average draw ratio of the film surface. The latter describes the strain distribution along the principal directions of the film plane. It is shown that $\bar{\lambda}$ and B depend on the final applied pressure and on the type of pressing surfaces used. The dependence of $\bar{\lambda}$ and B on the initial orientation and on the initial anisotropy of the precursors is also discussed in the light of structural arguments.

1 Introduction

It is well known that molecular orientation produced by uniaxial plastic deformation of polymers results in an enhancement of properties (elastic modulus, tensile strength, refractive index) in the direction parallel to the draw [1]. In case of films or sheets a biaxial orientation produces an enhancement of properties including tensile properties, toughness, clarity and shrinkability within the plane direction [2]. Orientation is imparted by thermomechanical treatments such as stretching at temperatures above the polymer glass transition and below its melt temperature [3]. Structure and properties of biaxially drawn materials have a high technological interest (particularly in the packaging industry) [4]. Biaxial orientation has been reported for blown and stretched (PE, PP, PET, polyamide) films [2, 5 to 9] for hydrostatically solid state extruded (PP) sheets [10, 11] for monoaxially drawn (PP and PET) films perpendicularly drawn to the initial drawing direction [12] and for biaxially drawn dried gels of ultra high molecular weight polyethylene [13].

The object of the present study is to describe a new method of processing polyethylene (PE) films by pressing uniaxially oriented samples. These samples were injection molded using molds that introduce axial elongational flow

during the filling process to obtain different degrees of initial uniaxial orientation. Specifically, the first part of the series (the present one) deals with the description and characterization of the strain in the three principal directions. Two functions are introduced whose study suffices to describe the system. The second part will report the mechanical properties of the produced biaxially oriented materials. The third part of the series will offer the study of their microstructure as revealed by wide and small angle X-ray scattering.

2 Experimental

Injection molded rods of PE, with a square cross section of $\sim 4 \times 4$ mm were produced following a method that introduces axial elongational flow during the filling process [14, 15]. The injection temperature was varied in order to achieve different degrees of orientation in the material. A number of injected samples using a conventional mold were also obtained for comparison. (Unless otherwise stated in what follows we will refer to "injection molding" as elongational flow injection molding.) Birefringence measurements, across the section of each rod, were made with a Leitz polarizing light microscope using a calcite compensator [15]. The material used in this investigation was linear PE, Vestolen A6012 (Hüls, Marl/Germany) with a viscosity average molecular weight of 113 000. From the point of view of mechanical properties and regarding its molecular weight this material can be considered as to lie within a brittle-ductile transition region [16]. Each injected rod was cut at fixed lengths and the segments were pressed between the plates of a hot press. Compression was always perpendicularly applied to the former injection flow. Forces between 30 and 600 kN were used. The pressing time was 20 s in all cases. Deformation was favored by continuous heating of the pressing plates to an optimum value of 127 ± 2 °C and by lubrication of the plates with silicon oil. The obtained PE films nearly have an elliptical shape and thicknesses ranging from 100 to 300 μ m depending on the mechanical treatment. The dimensions of the obtained films were measured after removing the pressure. In addition, to the compression of samples without any privileged direction (to be called uniaxially compressed samples), pressing plates with parallel carved 200 μ m wide grooves on their surface were also employed. The grooves on the upper and lower plates were kept parallel to each other and then

* Mail and present address: Dr. R. K. Bayer, Institut für Werkstofftechnik, University GH Kassel, Wilhelmshöher Allee 73, D-3500 Kassel, Germany.

alternatively placed along or perpendicular to the main axis of the moldings.

3 Definitions

The macroscopic strain behavior of the deformed samples can be characterized by three draw ratios λ_x , λ_y and λ_z defined along the three principal directions x , y and z (Fig. 1). Let us choose λ_x in the direction parallel to the former injection flow, λ_y to be normal to the flow direction and parallel to the film surface and λ_z normal to the thickness of the film. These three ratios are not independent of each other because of the existing relation between the volume before and after deformation. Hence, it was found more convenient to define two derived functions: biaxial draw ratio $\bar{\lambda}$ and degree of biaxiality B which will also account for a quantitative description of the problem. In the present

Fig. 1. Schematics of an injection molded sample before and after compression. Compression is exerted along the z -axis. The x -axis corresponds to the injection flow

work we have studied these magnitudes as a function of the final pressure applied to the samples. It will be shown that $\bar{\lambda}$ and B depend upon the following experimental conditions:

- Initial orientation of the sample, as defined by the mean birefringence Δn throughout the cross section. This orientation is a function of the melt temperature, T_m , during the injection molding of the precursors.
- Dimensions of the sample. The cross section has been kept constant and therefore the length of the sample x_0 is the effective parameter to be considered.
- Type of pressing surface: flat or with grooves.

3.1 Biaxial draw ratio

In what follows we shall use subindices "0" for the dimensions before compression and subindices "1" for the dimensions along compression (Fig. 1). The compression direction is along the z axis. In addition, the x -axis will be taken parallel to the initial uniaxial orientation axis (injection direction).

Let

$$\lambda_x = \frac{a_1}{a_0}; \quad \lambda_y = \frac{b_1}{b_0} \quad \text{and} \quad \lambda_z = \frac{c_1}{c_0}.$$

We then, define the biaxial draw ratio $\bar{\lambda}$, as the square root of the product of the draw ratios in the two principal directions parallel to the surface of the pressed film, i.e.,

$$\bar{\lambda} = (\lambda_x \lambda_y)^{1/2} \quad (1)$$

Fig. 2. Biaxial draw ratio as a function of final applied pressure for three samples ($\Delta n = 31 \cdot 10^{-3}$) having different a_0 . $\bar{\lambda}$ calculated according to Eqs. 1 (●) and 3 (△). Corrected values (Eq. 5) are represented by (▲)

In absence of any shape or volume change during compressive deformation it is known that:

$$\lambda_x \lambda_y \lambda_z = 1. \quad (2)$$

As a consequence the biaxial draw ratio will also be:

$$\bar{\lambda} = \lambda_z^{-1/2}. \quad (3)$$

A first inspection of the $\bar{\lambda}$ values obtained by means of Eqs. 1 and 3 is shown in Fig. 2, as function of P_F for three injection molded samples processed at $T_m = 143^\circ\text{C}$ ($\Delta n = 31 \times 10^{-3}$), pressed under a force of 400 kN. The different values of a_0 , yield the different final pressures P_F . Similar results can be found for all studied experimental conditions. It is obtained that $\bar{\lambda}$ calculated by means of Eq. 1 takes always a higher value than that derived from Eq. 3. The reason for this difference is that none of the two above assumptions is fully accomplished i.e., first the shape of the deformed moldings becomes elliptical and second an appreciable change in volume, ΔV , due to material deformation takes place. Hence, Eq. 2 must be modified in the following way:

$$\lambda_x \lambda_y \lambda_z = K \quad (4)$$

where K is a constant,

$$K = \frac{4}{\pi} \left(1 + \frac{\Delta V}{V_0} \right),$$

the first factor corresponding to the shape geometrical correction and

$$\frac{\Delta V}{V_0} = 1 - \frac{v}{0.5} \approx 0.2,$$

is derived from the Poisson's ratio for PE ($v \approx 0.4$). Using Eq. 4, we arrive at:

$$\bar{\lambda} = K^{1/2} \lambda_z^{-1/2}, \quad (5)$$

which now gives values in very good agreement with those calculated by means of Eq. 1 (see Fig. 2). In the particular

Fig. 3 (left). Final applied pressure as a function of the draw ratio in the two principal directions, parallel to the film surface (xy plane), of various samples having $A_0 = 1$ and $\Delta n \cdot 10^3 = 31$ ($\circ$); 21 ($\blacktriangle$); 9.4 (∇); 9.5 ($\square$). Isotropic sample ($\bullet$)

Fig. 4 (right). Final applied pressure as a function of the draw ratio in the two principal directions, parallel to the film surface (xy plane), of various injection moldings of the same material ($\Delta n = 21 \cdot 10^{-3}$) and different initial anisotropies: $A_0 = 1$ ($\blacktriangle$); 2.5 ($\square$); 7 ($\circ$) and 13 ($\blacksquare$)

case of an isotropic sample, with $a_0 = b_0$, the uniaxial compression can be considered as if it were an equibiaxial extension because in both cases $\lambda_x = \lambda_y = \bar{\lambda}$. In general, the biaxial draw ratio $\bar{\lambda}$ represents an average value of the draw ratio parallel to the film surface.

3.2 Degree of biaxiality

We define the degree of biaxiality simply as the quotient of the draw ratios λ_x and λ_y in the film plane:

$$B = \lambda_x / \lambda_y. \quad (6)$$

For $B = 1$, $\lambda_x = \lambda_y = \bar{\lambda}$ we have the case of an equilibrium extension. On the contrary, if $0 < B < 1$ then $\lambda_y > \lambda_x$ which in the limit can be considered as a state of predominant uniaxial deformation perpendicular to the former uniaxial orientation (injection direction). According to the definitions of Eqs. 5 and 6 the principal draw ratios can now be expressed in terms of both parameters, $\bar{\lambda}$ and B :

$$\begin{aligned} \lambda_x &= \bar{\lambda} B^{1/2}, \\ \lambda_y &= \bar{\lambda} B^{-1/2}, \\ \lambda_z &= K \bar{\lambda}^{-2}. \end{aligned} \quad (7)$$

4 Results and discussion

4.1 The influence of orientation and anisotropy on the degree of biaxiality

4.1.1 Flat pressing surfaces

Let us first examine the case of injected samples with $a_0 = b_0$ subjected to compression without any privileged direction. Fig. 3 shows the increasing individual dependence of, both, λ_x and λ_y upon the final applied pressure P_F . Pressure has been taken as ordinate in order to refer to compressive stress-strain plots. If the shape anisotropy A is defined as (Fig. 1):

$$A = a/b, \quad (8)$$

for all samples in Fig. 3 $A_0 = 1$ ($a_0 = b_0$). The mean birefringence, Δn , is taken here as a parameter. On the contrary

in Fig. 4, $\Delta n = 21 \times 10^{-3}$ ($T_m = 170^\circ\text{C}$) is constant and A_0 is taken now as parameter. From Figs 3 and 4 it is seen that for each fixed pair of variables ($A_0, \Delta n$), the two corresponding curves $\lambda_x(P_F)$ and $\lambda_y(P_F)$ are increasing functions of P_F . For $P_F = 0$, $\lambda \rightarrow 1$. In addition, it is observed that in each case $\lambda_y \geq \lambda_x$. For $\lambda_y = \lambda_x$ we have the case of the isotropic material with $A_0 = 1$ (Fig. 3). Furthermore, for any value $A_0 = 1$ the biaxiality $B = \lambda_x / \lambda_y$ decreases with increasing prior orientation (decreasing T_m). Similarly, for a given material, B decreases with increasing values of the shape anisotropy A_0 (Fig. 4).

From the foregoing it is evident that deformation along the two principal directions, parallel to the surface, can be monitored between both limiting values of B by means of an adequate variation of the two parameters A_0 and Δn . Another way to analyze the obtained results is by plotting the final anisotropy A_1 against the initial anisotropy A_0 for different values of Δn and P_F (Figs. 5 and 6). In all cases, irrespective of the type of pressing surfaces used, an empirical law of the type:

$$A_1 = \mu A_0^{1/3}, \quad (9)$$

has been found where μ is a parameter which is a function of Δn . Using Eqs. 6 and 9, Eq. 10 can be rewritten as:

$$B = \mu A_0^{-2/3}, \quad (10)$$

showing that biaxiality is a decreasing function of the initial anisotropy, and that B also depends on μ . As it can be inferred from Fig. 7 for various initial anisotropies and a fixed $\Delta n = 21 \times 10^{-3}$, the final pressure does not significantly influence the biaxiality B , in the range of investigated pressures. An isotropic PE sample with $A_0 = 1$ has also been included for comparison. These results emphasize the fact that when P_F increases, inducing, as a result, an increase in the lateral dimensions of the film represented through $\bar{\lambda}$, biaxiality B remains constant. Hence from Eq. 10 it turns out that neither μ depends on P_F . By using Eqs. 10 and 7 we arrive at

$$\lambda_y = \mu^{-1/2} A_0^{1/3} \bar{\lambda}, \quad \lambda_x = \mu^{1/2} A_0^{-1/3} \bar{\lambda}, \quad (11)$$

Fig. 5 (left). Anisotropy of the uniaxially compressed samples against initial anisotropy. Symbols have the same meaning as in Fig. 3. The solid line corresponds to the predicted isotropic behavior

Fig. 6 (right). Anisotropy of the deformed samples, using pressing plates with grooves, against initial anisotropy. Injection moldings: – elongational flow: $\overline{\Delta n} = 31 \cdot 10^{-3}$ ($\bullet$, $\circ$); $9 \cdot 10^{-3}$ ($\blacksquare$, $\square$) – conventional mold: $\overline{\Delta n} = 6 \cdot 10^{-3}$ ($\blacktriangle$, $\triangle$) Grooves are parallel (open symbols) or perpendicular (solid symbols) to the injection direction. Curves 1, 2 and 3 describe the behavior of isotropic samples (see text)

which analytically justifies the results of Figs. 3 and 4. The draw ratios λ_y and λ_x in the two principal directions parallel to the surface of the films are proportional to the biaxial deformation $\bar{\lambda}$ modulated by two factors: the first one (μ) depends on the orientational state of the material character-

ized by $\overline{\Delta n}$ and on the pressing conditions. The second one, A_0 , is a function of the initial anisotropy of the samples. Table 1 collects the μ -values derived from Fig. 5. The value of $\mu = 1$ corresponds to the deformation of isotropic samples and their behavior is represented by the solid line in Fig. 5. It is noteworthy that the experimental value of a compression molded PE isotropic sample ($A_0 = 1, A_1 = 1$)

Table 1. Injection melt temperature, mean birefringence and μ -values for uniaxially compressed PE moldings

T_m °C	$\overline{\Delta n} \times 10^3$	μ
143	31	0.50
170	21	0.70
257	9.5	0.80
331	9.5	0.96
Isotropic	0	1

effectively lies on the predicted line. For the injection molded samples μ concurrently decreases with increasing mean birefringence which characterizes the initial material orientation.

4.1.2 Pressing surfaces with grooves

The influence of pressing plates with grooves on the μ -values is illustrated in Table 2. We have considered two specific alternatives: grooves parallel to the injection direction (suffix: $\parallel$) and perpendicular to it (suffix: $\perp$). Evidently

Fig. 7. Degree of biaxiality as a function of final applied pressure for the same material ($\overline{\Delta n} = 21 \cdot 10^{-3}$) with different initial anisotropies $A_0 = 1$ ($\blacktriangle$); 2.3 ($\square$); 7 ($\circ$) and 13 ($\blacksquare$). An isotropic sample with $A_0 = 1$ has also been included ($\bullet$)

Table 2. Injection melt temperature, mean birefringence and μ -values of compressed PE moldings, using pressing plates with grooves placed parallel ($\parallel$) or perpendicular ($\perp$) to the previous injection flow. C: conventional mold

T_m °C	$\Delta n \times 10^3$	$\mu_{\parallel}$	$\mu_{\perp}$	$\bar{\mu} = \sqrt{\mu_{\parallel}\mu_{\perp}}$
143	31	0.76	0.57	0.66
170	21	0.83	0.65	0.73
257	9.5	1.03	0.70	0.85
298	9	1.05	0.75	0.89
331	9.5	1.10	0.80	0.94
259(C)	6	1.10	0.81	0.94

guides will always favour material flow in their own direction at expenses of the perpendicular direction. It is found, that the biaxial draw ratio $\bar{\lambda}$ as well as the principal draw ratios λ_x and λ_y , are always smaller than the corresponding values in simple uniaxial compression. This is a consequence of the increase in the friction (specific surface) introduced by the grooves. The following relations can, thus, be written:

$$\begin{aligned}\lambda_x &> \lambda_{x\parallel} > \lambda_{x\perp}, \\ \lambda_y &> \lambda_{y\perp} > \lambda_{y\parallel},\end{aligned}\quad (12)$$

which according to the experimental results yield:

$$\bar{\lambda} > \bar{\lambda}_{\parallel} \approx \bar{\lambda}_{\perp}, \quad (13)$$

In addition biaxiality B can also be modified by the use of grooved plates. According to Table 2:

$$\mu_{\parallel} > \mu > \mu_{\perp}, \quad (14)$$

and from Eq. 11, we arrive at:

$$B_{\parallel} > B > B_{\perp}, \quad (15)$$

Eq. 15 means that biaxiality is favoured when grooves are placed in the same direction as the former uniaxial orientation axis of the injected PE. In this case grooves tend to compensate the hindrance to deformation introduced by the previous orientation. The mean value $\bar{\mu} = \sqrt{\mu_{\parallel}\mu_{\perp}}$ obtained for compressed materials, using pressing plates with grooves, has also been included in Table 2. This value does not coincide with the μ values of Table 1 derived for uniaxially compressed samples because of the different pressing conditions. In Fig. 6, the two solid lines 2 and 3 represent the deformation modes of isotropic PE samples pressed with grooved plates. Grooves are here respectively placed parallel (curve 2) and perpendicular (curve 3) to the x axis. Finally, the case of flat pressing plates is also shown for comparison (curve 1).

4.2 Interpretation of the empirical expression for biaxiality

In the foregoing (Eq. 10) we have shown that biaxiality B is a function of, both, the initial anisotropy and orientation, the latter being described by the parameter μ . Let us try to justify this double dependence. On the one hand, the morphology of the injection molded rods used as starting materials is known to be one of shish-kebab highly rigid fibrils oriented in the injection direction [16]. Thus, on compression

of the preoriented sample, it is to be expected that the macroscopic strain should preferably develop in the y-interfibrillar direction, perpendicular to the former orientation direction. In terms of our description this would lead to a decrease of B. This behaviour is well described by Eq. 10 which emphasizes a direct proportionality between B and μ , and takes into account that μ is a decreasing function of initial orientation (Tables 1 and 2).

The B-dependence on the initial anisotropy of the samples is, on the other hand, somewhat more complex to explain. A dependence of the local deformation on the initial dimensions of the sample has been earlier reported in case of uniaxial drawing of thermoplastic polymers by Springer et al. [17]. These authors show that a relationship of the type:

$$\lambda \sim a_0^{k_1} b_0^{-k_2}, \quad (16)$$

holds, where the initial dimensions a_0 and b_0 are taken independently from each other and k_1 and k_2 are two arbitrary exponents. However, an explanation for this behavior is still missing. One may attempt to interpret the dependence of λ_x on b_0 in the light of the transformation of the initial isotropic structure into the final highly oriented fiber structure occurring behind the neck [18]. According to this concept, the destruction zones, separating the new microfibrils from the not yet transformed stacks of original lamellae, are much smaller (a few hundred nm) than the lateral dimensions of the sample. One may argue that a reduction of sample thickness b_0 would result in a decrease of the number of destructive zones. This would produce, in turn, an enhanced plastic compliance giving rise to an improvement of chain orientation, i.e. a larger drawability.

In the case of a macroscopic compressive strain a non-homogeneous distribution of the deformation has been also observed [19]. It is noteworthy that our experimental results on preoriented materials show that λ_x is inversely proportional to a_0 . If we now assume that a_0 and b_0 are inversely related as in the case of uniaxial drawing (in our experiments b_0 was kept constant), then we can write:

$$\lambda_x \sim a_0^{-k_2} b_0^{k_1}. \quad (17)$$

In the perpendicular direction, we would also have:

$$\lambda_y \sim a_0^{k_1} b_0^{-k_2}. \quad (18)$$

By division of Eqs. 17 and 18 one obtains B:

$$B \sim a_0^{-K} b_0^K = A_0^{-K} \quad (19)$$

where $K = k_1 + k_2$; expression which is based on Springer's observations and which, if we let $K = 2/3$, justifies the above empirical equation found for the biaxiality.

4.3 Dependence of the biaxial draw ratio on pressure and biaxiality

The biaxial draw ratio $\bar{\lambda}$ depends not only on the applied pressure but also on the orientation of the samples before pressing as it is shown in Fig. 8. It can be observed that biaxial draw ratio increases with decreasing orientation, i.e. with increasing μ . In this figure A_0 is kept constant ($A_0 = 1$). Fig. 9 depicts the biaxial draw ratio $\bar{\lambda}$ as a

Fig. 8. Final applied pressure versus biaxial draw ratio for two samples with $A_0 = 1$: i) isotropic (●), ii) injection molded ($\Delta n = 21 \cdot 10^{-3}$) (▲)

Fig. 9. Final applied pressure versus biaxial draw ratio for the material with $\Delta n = 21 \cdot 10^{-3}$ with different initial anisometries. Symbols have the same meaning as in Fig. 4

Fig. 10. Final applied pressure versus biaxial draw ratio for the material with $\Delta n = 9.4 \cdot 10^{-3}$ and two different anisometries. $A_0 = 1$ (▲) and $A_0 = 13$ (■)

function of P_F , for a fixed ($\Delta n = 21 \cdot 10^{-3}$) and various initial anisometries A_0 . The obtained data seem to be segregated according to their A_0 values. To confirm this tendency, a new material with ($\Delta n = 9.4 \cdot 10^{-3}$) has been investigated for two widely different anisometries namely $A_0 = 1$ and $A_0 = 12$, in the lower range of final applied pressures (Fig. 10). From Figs. 9 and 10 it becomes clear that A_0 (initial shape of the sample) has an influence on $\bar{\lambda}$, as it was previously shown on B (Eq. 10). From both experimental facts it may be postulated a certain relation between $\bar{\lambda}$ and B. This means that the larger the biaxiality B is, the greater is the increase of $\bar{\lambda}$ when a given pressure P_F is applied. When B tends to zero (uniaxial deformation), $\bar{\lambda}$ decreases, whereas in the case of equibiaxial deformation, $\bar{\lambda}$ achieves its maximum value.

A description of the biaxial draw ratio (compressive stress-strain plot) in terms of van der Waals theory of real networks [20] will be given in a separate study.

5 Conclusions

The main results of this work can be summarized as follows.

A quantitative description of the strain behaviour of biaxial films can be achieved by introducing two magnitudes: biaxial draw ratio ($\bar{\lambda}$) and degree of biaxiality (B). The draw ratio in the three principal directions can be derived from $\bar{\lambda}$ and B (Eq. 8).

For pressed injection molded materials both, B and $\bar{\lambda}$, depend upon the orientational state of the starting material which is characterized by the mean birefringence (Δn). B and $\bar{\lambda}$ can also be modified by changing the dimensions of the PE precursors.

The final applied pressure to the oriented material only influences the biaxial draw ratio $\bar{\lambda}$.

For compression using pressing plates with grooves, $\bar{\lambda}$ is lower than for simple uniaxial compression irrespective of the relative orientation of the grooves. On the other hand B

can be monitored, by means of the relative orientation of the grooved pressing surfaces leading to values which can be higher or lower than those obtained for uniaxial compression.

References

- 1 Peterlin, A.: Colloid Polym. Sci. 265, p. 357 (1987)
- 2 Siegmund, A., Niv, Y.: Polym. Eng. Sci. 27, p. 225 (1987)
- 3 Hensen, F.: Kunststoffe 71, p. 643 (1981)
- 4 Benning, C. J.: Plastic Films for Packaging. Technomic Pub. Co, New York 1983
- 5 Nagasawa T., Maslumura, T., Hoshimo, S.: Appl. Polym. Symp. 20, p. 275 (1975)
- 6 De Vries A. J., Bonnebat, C., Beutemps, J.: J. Polym. Sci. Polym. Symp. 58, p. 109 (1977)
- 7 Maddams, W. F., Preedy, J. E.: J. Appl. Polym. Sci. 22, p. 2721 (1987)
- 8 Choi, K. J., Spruiell, J. E., White, J. L.: J. Polym. Sci. Polym. Phys. Ed. 20, p. 27 (1982)
- 9 Kohno, M., Tamemoto, K.: Polym. Eng. Sci. 27, p. 558 (1987)
- 10 Pan, S. J., Tang, H. J., Hiltner, A., Baer, E.: Polym. Eng. Sci. 27, p. 869 (1987)
- 11 Tang, H. J., Hiltner, A., Baer, E.: Polym. Eng. Sci. 27, p. 876 (1987)
- 12 Hinrichsen, G., Eberhardt, A., Lippe, U., Springer, H.: Colloid Polym. Sci. 259, p. 73 (1981)
- 13 Sakai, Y., Miyasaka, K.: Polymer 29, p. 1608 (1988)
- 14 Bayer, R. K., Zachmann, H. G., Baltá Calleja, F. J., Umbach, H.: Polym. Eng. Sci. 29, p. 186 (1989)
- 15 López Cabarcos, E., Bayer, R. K., Zachmann, H. G., Baltá Calleja, F. J., Meins, W.: Polym. Eng. Sci. 29, p. 193 (1989)
- 16 Bayer, R. K., Baltá Calleja, F. J., López Cabarcos, E., Zachmann, H. G., Paulsen, A., Brüning, F., Meins, W.: J. Materials Sci. 24, p. 2643 (1989)
- 17 Springer, H., Jäger, H. G., Hinrichsen, G.: Colloid Polym. Sci. 258, p. 227 (1980)
- 18 Peterlin, A., Baltá Calleja, F. J.: J. Appl. Phys. 40, p. 4238 (1969)
- 19 Peterlin, A.: Kolloid Z.Z. Polymere 233, p. 857 (1969)
- 20 Kilian, H. G.: Polymer, 22, p. 209 (1981)

Date received: August 18, 1990

Date accepted: September 24, 1990