Journal of Engineering Physics and Thermophysics, Vol. 83, No. 5, 2010

DEVELOPMENT OF AN IDENTIFICATION SYSTEM FOR BIAXIALLY ORIENTED POLYMER FILMS BASED ON THE DEGREE OF THEIR TRANSVERSE EXTENSION

V. V. Goncharenko, I. O. Mikulenok, D. G. Shvachko, and D. N. Shved

UDC 678.01:539.3

An expression has been developed that generalizes three basic geometric schemes of film deformation (axial, planar, and biaxial extension) and also all intermediate schemes. The system of quantitative identification of nonuniformly biaxially oriented films according to their transverse extension has been tested on six different film types.

Keywords: *polymer, film, biaxial extension, orientation, identification.*

Introduction. Biaxially oriented polymer films include a very wide class of films whose degree of extension ranges fairly broadly. Uniaxially oriented (extended) polymer films are produced in the limiting case with no transverse extension. In the case where polymer films are subjected to the same extension in longitudinal and transverse directions, they become uniformly biaxially oriented (extended). Finally, when the degree of transverse extension of the film is half as large as its longitudinal extension, a planar film orientation is realized.

In fact, deformation of polymer films by the basic schemes described above results in nonuniformly biaxially oriented films which more or less correspond in the degree of transverse extension to one of the indicated cases. The degree of transverse thermal shrinkage of such films can vary within fairly wide limits (from negative values to values larger than the degree of longitudinal thermal shrinkage).

Since the study of any object necessitates its identification, in order to locate the place occupied by a biaxially oriented film of specific type and dimensions in the above class of films, one should have a clear system of its identification which would unambiguously determine this place quantitatively. The development of the identification system can be regarded as the first step toward systematic technological investigations of nonuniformly biaxially oriented polymer films. Since the industrial branch producing a wide range of biaxially oriented films is characterized by high dynamics of the development, working out a system of technological identification of such films is a very topical problem.

The wide class of nonuniformly biaxially oriented films is limited, on the one hand, by uniaxially oriented films, and on the other, by films in which the transverse deformation exceeds the longitudinal one. To make it possible to identify such films according to the degree of transverse extension, it is necessary to obtain a mathematical expression generalizing their properties, or more exactly, a rheological equation of their state. The current study aims at solving such a problem within the framework of the linear theory of viscoplasticity [1].

Three Deformation Schemes. The most convenient method for assessing the degree of orientation of biaxially oriented polymer films is the method of thermal shrinkage. Such shrinkage is effected at a temperature 5–10°C higher than the melting temperature of polymer crystals. Here the specimen becomes devoid of any loads preventing free restoration of the accumulated highly elastic deformations. The degree of longitudinal thermal shrinkage S_1 is here defined as

$$S_1 = (l_0/l_t - 1)\cdot 100\% \,. \tag{1}$$

The degree of transverse thermal shrinkage S_2 is determined from the expression

---

Ukrainian Technical University "Kiev Polytechnic Institute," 30 Pobeda Ave., Kiev, 03056, Ukraine; email: vvgonch@ukr.net. Translated from Inzhenerno-Fizicheskii Zhurnal, Vol. 83, No. 5, pp. 949–953, September–October, 2010. Original article submitted March 3, 2009; revision submitted December 18, 2009.

 1062-0125/10/8305-1010

TABLE 1. Stress Tensors and Deviators

Parameters	Deformation schemes		
	A	P [2]	B
Stress tensor	$\sigma_{ij} = \left\lvert\begin{matrix} \sigma_E & & \\ & 0 & \\ & & 0 \end{matrix}\right\rvert$	$\sigma_{ij} = \left\lvert\begin{matrix} \sigma_P & & \\ & 0.5\sigma_P & \\ & & 0 \end{matrix}\right\rvert$	$\sigma_{ij} = \left\lvert\begin{matrix} \sigma_B & & \\ & \sigma_B & \\ & & 0 \end{matrix}\right\rvert$
Stress deviator	$\sigma'_{ij} = \frac{2}{3}\sigma_E \left\lvert\begin{matrix} 1 & & \\ & -0.5 & \\ & & -0.5 \end{matrix}\right\rvert$	$\sigma'_{ij} = \frac{\sigma_P}{2} \left\lvert\begin{matrix} 1 & & \\ & 0 & \\ & & -1 \end{matrix}\right\rvert$	$\sigma'_{ij} = \frac{\sigma_B}{3} \left\lvert\begin{matrix} 1 & & \\ & 1 & \\ & & -2 \end{matrix}\right\rvert$
Factor of transverse extension	$\zeta = 0$	$\zeta = 0.5$	$\zeta = 1$

TABLE 2. Deviators ε^e_{ij} and $\dot{\varepsilon}_{ij}$

Deviators	Deformation schemes		
	A	P	B
Elastic deformation	$\varepsilon^e_{ij} = \varepsilon^e_1 \left\lvert\begin{matrix} 1 & & \\ & -0.5 & \\ & & -0.5 \end{matrix}\right\rvert$	$\varepsilon^e_{ij} = \varepsilon^e_1 \left\lvert\begin{matrix} 1 & & \\ & 0 & \\ & & -1 \end{matrix}\right\rvert$	$\varepsilon^e_{ij} = \varepsilon^e_1 \left\lvert\begin{matrix} 1 & & \\ & 1 & \\ & & -2 \end{matrix}\right\rvert$
Deformation rate	$\dot{\varepsilon}_{ij} = \dot{\varepsilon}_1 \left\lvert\begin{matrix} 1 & & \\ & -0.5 & \\ & & -0.5 \end{matrix}\right\rvert$	$\dot{\varepsilon}_{ij} = \dot{\varepsilon}_1 \left\lvert\begin{matrix} 1 & & \\ & 0 & \\ & & -1 \end{matrix}\right\rvert$	$\dot{\varepsilon}_{ij} = \dot{\varepsilon}_1 \left\lvert\begin{matrix} 1 & & \\ & 1 & \\ & & -2 \end{matrix}\right\rvert$

TABLE 3. Values of Elasticity Moduli and Viscosity for Three Deformation Schemes

Parameters	Deformation schemes		
	A	P	B
Elasticity moduli	$E_E = 3G$	$E_P = 4G$	$E_B = 6G$
Viscosity	$\eta_E = 3\eta$	$\eta_P = 4\eta$	$\eta_B = 6\eta$
Factor of transverse extension	$\zeta = 0$	$\zeta = 0.5$	$\zeta = 1$

$$S_2 = (b_0/b_t - 1)\cdot 100\% \,. \tag{2}$$

The ratios l_0/l_t and b_0/b_t can be considered as principal degrees of elastic deformation of the specimen (in the longitudinal λ^e_1 and transverse λ^e_2 directions) that is accumulated in the film forming. For uniformly biaxially oriented films, $S_1 = S_2$ and $\lambda^e_1 = \lambda^e_2$. For polymer films produced in the case of planar extension, the degree of transverse shrinkage is zero ($S_2 = 0$ and $\lambda^e_2 = 1$). The transverse shrinkage for uniaxially oriented films is negative, since the film reduction in the longitudinal direction involves its transverse expansion.

Table 1 presents the stress tensor σ_{ij} and its deviator component σ'_{ij} for the three most studied geometric schemes and the factor of transverse extension $\zeta = \sigma_2/\sigma_1$ (σ_1 and σ_2 are the longitudinal and transverse principal stresses, respectively).

Deviator components of the elastic-deformation tensor ε^e_{ij} and of the deformation-rate tensor $\dot{\varepsilon}_{ij}$ for three basic deformation schemes are supplied in Table 2.

According to the linear theory of viscoplasticity [1], the elastic mechanical behavior of polymer systems is described by Hooke's law in tensor form:

$$\sigma'_{ij} = 2G\varepsilon^{e}_{ij}\,, \tag{3}$$

and a viscous flow is described by Newton's law in tensor form:

$$\sigma'_{ij} = 2\eta\dot{\varepsilon}_{ij}\,. \tag{4}$$

Substituting the deviator parts of stress tensors (Table 1) and of elastic-deformation and deformation-rate tensors (Table 2) into expressions (3) and (4) gives the values of the moduli of elasticity E_E, E_P, and E_B and longitudinal viscosities η_E, η_P, and η_B respectively for the three geometric schemes of deformation A, P, and B [1, 2] presented in Table 3.

Unification of Three Deformation Schemes. The above systematization of results based on the linear theory of viscoplasticity allows one to develop a single expression that unifies the three basic geometric schemes of deformation. In study [3], such a correlation is proposed in the form of a deviator component of the deformation-rate tensor:

$$\dot{\varepsilon}_{ij} = \varepsilon_0 \begin{vmatrix} 1 & 0 & 0 \\ 0 & m & 0 \\ 0 & 0 & -(1+m) \end{vmatrix}. \tag{5}$$

Indeed, at $m = -0.5$ deviator (5) corresponds to the axial extension (scheme A), at $m = 0$, to the planar stretching (scheme P), and at $m = 1$, to the uniform biaxial extension (scheme B). This approach to calculating the determining factor precludes the unification of all known geometric schemes of deformation by a single expression.

The simplest solution of this problem is suggested in Table 1: if use is made of the factor of transverse extension ζ, which is defined as the ratio of the transverse principal stress σ_2 to its longitudinal value σ_1, the stress tensor for basic (A, P, and B) and intermediate geometric schemes of deformation corresponding to the nonuniform biaxial film extension can be represented as

$$\sigma_{ij} = \sigma_0 \begin{vmatrix} 1 & & \\ & \zeta & \\ & & 0 \end{vmatrix}. \tag{6}$$

The deviator part of this tensor in this case takes the form

$$\sigma'_{ij} = \frac{1}{3}\sigma_0 \begin{vmatrix} 2-\zeta & & \\ & -1+2\zeta & \\ & & -1-\zeta \end{vmatrix}. \tag{7}$$

In accordance with deviator (7), all deviators presented in Table 2 and all cases of the nonuniform biaxial stretching are described by expressions

$$\varepsilon^{e}_{ij} = \frac{1}{2-\zeta}\varepsilon^{e}_{1} \begin{vmatrix} 2-\zeta & & \\ & -1+2\zeta & \\ & & -1-\zeta \end{vmatrix}, \tag{8}$$

$$\dot{\varepsilon}_{ij} = \frac{1}{2-\zeta}\dot{\varepsilon}_{1} \begin{vmatrix} 2-\zeta & & \\ & -1+2\zeta & \\ & & -1-\zeta \end{vmatrix}. \tag{9}$$

There is no difficulty in convincing ourselves that, at the degrees of transverse extension presented for the three deformation schemes in Table 1, expressions (7)–(9) substituted in relations (3) and (4) give the values of the moduli of elasticity

$$E(\zeta) = 6G\frac{1}{2-\zeta}\,, \tag{10}$$

TABLE 4. Experimental Data for Five Industrial Processes and One Laboratory (III) Process of Production of Sleeve Polyethylene Films

Film Nos.	A, mm	δ, mm	v, cm/s	δ_0, mm	d, mm	T, °C	l_0/l_t, mm	b_0/b_t, mm
I	123	0.045	60	0.6	40	190	$\frac{100}{66.7}$	$\frac{50}{28.6}$
II	380	0.033	40	0.7	90	170	$\frac{100}{17}$	$\frac{50}{24}$
III	182	0.065	47	1.1	64	180	$\frac{100}{60.6}$	$\frac{50}{45}$
IV	44	0.0625	26.8	1.0	30	170	$\frac{100}{14.5}$	$\frac{50}{53}$
V	460	0.0156	83.6	0.7	90	170	$\frac{100}{14.3}$	$\frac{50}{23}$
VI	34.5	0.081	26.8	1.0	30	170	$\frac{100}{18}$	$\frac{70}{90}$

TABLE 5. Results of Processing of Experimental Data

Calculating equation	Film Nos.					
	I	II	III	IV	V	VI
$D = 2A/\pi$	78.3	242	116	28	293	21.9
$q = 2A\delta v$	5.12	10	1.11	1.5	11.9	1.5
$v = q(\pi d\delta_0)^{-1}$	6.8	5.05	0.537	1.6	6.0	1.6
$\varepsilon_1^e = \ln (l_0/l_t)$	0.4	1.77	0.5	1.9	1.95	1.7
$\varepsilon_2^e = \ln (b_0/b_t)$	0.56	0.742	0.09	–0.06	0.78	–0.25
$\frac{2\zeta - 1}{2 - \zeta} = \frac{\varepsilon_2^e}{\varepsilon_1^e} = k$	1.4	0.419	0.18	–0.03	0.4	–0.15
$\zeta = \frac{2k+1}{2+k}$ (for $\zeta > 0.5$)	1.12	0.76	0.624	0.477	0.75	0.38
B = 2(ζ – 0.5)·100% (for $\zeta < 0.5$)	124	52	24.8	—	50	—
P = 2 ζ ·100% (for $\zeta > 0.5$)	—	—	—	95.4	—	76
P = (100 – B) (for $\zeta < 0.5$)	—	48	75.2	—	50	—
A = (100 – B)	0	0	0	5.6	—	24

and viscosity

$$\eta (\zeta) = 6\eta \frac{1}{2 - \zeta}, \tag{11}$$

supplied in Table 3.

Thus, within the framework of the linear theory of viscoelasticity, using Eqs. (10) and (11) it is possible to calculate the moduli of elasticity $E(\zeta)$ and viscosity $\eta(\zeta)$ for all intermediate geometric schemes of deformation corresponding to the nonuniform biaxial extension.

Experiment. The identification system developed was tested in practice using experimental data for five industrial processes and one laboratory (III) process of production of biaxially oriented sleeve polymer films presented in Table 4. Calculated results for the degree of identity of polymer films with respect to the transverse stretching are given in Table 5. The factor of transverse ζ extension was determined by the method of thermal shrinkage of the considered polymer films using the equation derived from expression (8):

$$(2\zeta - 1)/(2 - \zeta) = [\ln (b_0/b_t)]/[\ln (l_0/l_t)] . \tag{12}$$

Nonuniformly biaxially oriented polymer films were divided into two classes according to the degree of transverse extension. For the first class of polymer films with $\zeta > 0.5$, we initially determined the degree of uniformity B of the biaxial extension (more exactly, the degree of identity to the uniform biaxial stretching):

$$\mathrm{B} = 2\,(\zeta - 0.5)\cdot 100\% , \tag{13}$$

and then the degree of identity P with the planar extension:

$$\mathrm{P} = (100 - \mathrm{B}) . \tag{14}$$

For the second class of polymer films with $\zeta < 0.5$, we first determined the degree of identity with the planar extension:

$$\mathrm{P} = 2\zeta\cdot 100\% \tag{15}$$

and then the degree of identity with the axial extension:

$$\mathrm{A} = (100 - \mathrm{P}) . \tag{16}$$

Conclusions. Proceeding from the linear theory of viscoelasticity we developed a generalized system of calculating components of deviator parts of the stress, elastic-deformation, and deformation-rate tensors based on the degree of transverse extension for all geometric schemes of deformation implemented in the production of biaxially oriented polymer films. The developed system for determining the deviator components is applicable for three deformation cases: uniaxial stretching, uniform biaxial stretching, and planar extension. Relying on the generalized system of calculating components of the elastic-deformation deviator, a system is worked out for quantitative identification of nonuniformly biaxially oriented actual polymer films whose degree of transverse thermal shrinkage changes from negative to positive values.

NOTATION

A, width of a folded sleeve, mm; b_0 and b_t, specimen widths before and after thermal shrinkage, respectively, m; *D*, diameter of a film sleeve, mm; *d*, annulus diameter, mm; E_E, E_P, and E_B, longitudinal elasticity moduli of the uniaxial, uniform biaxial, and planar extension, respectively, Pa; *G*, elasticity modulus in shear, Pa; *k*, factor of transverse thermal shrinkage; l_0 and l_t, specimen lengths before and after thermal shrinkage, respectively, m; *m*, index of nonuniformity of the biaxial extension; *q*, volumetric productivity, cm^3/s; S_1 and S_2, degrees of longitudinal and transverse thermal shrinkage, respectively; *T*, melt temperature, $^{\mathrm{o}}$C; *v*, rate of the film output, cm/s; δ, film thickness, mm; δ_0, annulus thickness, mm; $\varepsilon_{ij}^{\mathrm{e}}$ and $\dot{\varepsilon}_{ij}$, deviators of elastic deformation and deformation rate, respectively, s^{-1}; $\varepsilon_1^{\mathrm{e}}$ and $\varepsilon_2^{\mathrm{e}}$, principal longitudinal and transverse elastic deformation, respectively; $\dot{\varepsilon}_1$, principal longitudinal velocity of deformation, s^{-1}; ζ, factor of transverse extension at arbitrary stretching; ζ_E, ζ_P, and ζ_B, factors of transverse extension in the axial, planar, and uniform biaxial stretching, respectively; η, viscosity, Pa·s; η_E, η_P, and η_B, longitudinal viscosities in the uniaxial, planar, and uniform biaxial stretching, respectively, Pa·s; λ_1^{e} and λ_2^{e}, principal degrees of elastic deformation of the specimen in longitudinal and transverse directions, respectively; σ_{ij} and σ'_{ij}, stress tensor and the stress deviator, respectively, Pa; σ_E, σ_P, and σ_B, principal longitudinal stresses in the uniaxial, planar, and uniform biaxial stretching, respectively, Pa; σ_0, component of the stress tensor in principal axes, Pa. Subscripts and superscripts: E, elongation; P, planar; B, biaxial; e, elastic; i = 1, 2, 3 and j = 1, 2, 3, serial indices; t, thermal; 0, initial state; 1 and 2, longitudinal and transverse direction of the extension, respectively.

REFERENCES

1. A. Ya. Malkin and A. I. Isayev, *Rheology. Conceptions. Methods. Applications*, Toronto, Canada: Chem. Tech. Publishing (2006).
2. M. Takahashi, T. Isaki, T. Takigawa, and T. Masuda, Measurement of biaxial and uniaxial extensional flow behavior of polymer melts at constant strain rates, *J. Rheol.*, **37**, No. 5, 827–846 (1993).
3. T. Raible, S. E. Stephenson, J. Meissner, and M. H. Wagner, Constant force elongation flow of low-density polyethylene melt — experiment and theory, *J. Non-Newton. Fluid Mech.*, **11**, No. 3, 239–254 (1982).