

Investigation of the elastic-viscous-plastic behavior of thermoplastic materials. Experiment and simulation

Oleg K. Garishin Anton S. Korlyakov Vladimir V. Shadrin
shadrin@icmm.ru

Abstract

Thermoplastics are polyolefin polymer materials which on heating are able to transfer reversibly into a high-elasticity state and then into a plastic state. Such a behavior of these polymers permits molding a wide variety of products. Polyethylene is a main industrial product of this class of materials. It has a complex mechanical behavior and demonstrates the well-defined elastoplastic and viscoelastic properties. This work is devoted to the experimental and theoretical study of these properties. In order to obtain in a single experiment all necessary experimental data on the elastic-visco-plastic behavior of the polymer, an appropriate procedure based on the cyclic deformation of sample has been developed which includes: stretching — stress relaxation — reducing strain to some predetermined constant value of tensile strength — stress relaxation — next cycle of deformation. Each subsequent cycle is made with increasing strain amplitude.

The model is based on a differential approach to the construction of the constitutive equations of mechanical behavior of materials with the help of symbolic schemes. The approach uses a mathematical apparatus of mechanics of nonlinear finite deformations. The symbolic model scheme consists of two parallel branches containing two serially connected elements: a) elastic and plastic, b) elastic and viscous. The elastoplastic branch represents the behavior of agglomerates of more rigid crystallites, their displacement and destruction during deformation. The viscoelastic branch describes the flow of the amorphous polymer phase.

The experimental data obtained for polyethylene PE 107-02K were theoretically analyzed using this model. Dependencies of elastic, plastic and viscous model parameters on polymer deformation were obtained. A comparison of the calculated and experimental results shows high coincidence. This is indicative of the fact that the conclusions drawn from the analysis of model parameters are close to reality.

Thermoplastics are polyolefin polymer materials which on heating are able to transfer reversibly into a high-elasticity state and then into a plastic state. This permits molding a wide variety of products.[1]. Polyethylene (PE) holds today the first place in industrial production (both in a pure form and with a wide variety of fillers). There are many different types of polyethylene with different molecular weight (polymer molecule length), branching level (number of pendant groups), crystallinity degree (a share of molecular chains arranged in an orderly fashion — crystallites — in the total volume of the material) [2]. Accordingly, their physical and mechanical properties are also different. For example, the elastic modulus of isotropic PE varies from 100 MPa to 1000 MPa, and the strength may vary tenfold. Common to all types of polyethylene is that they are characterized by a complex mechanical behavior and show the well-defined non-linear elastic, viscous and plastic properties.

The experimental investigation of polyethylene under finite strains is a complex and time-consuming task, since the viscoelastic and elastoplastic characteristics of the material

change during loading. The idea of using different variants of cyclic tests to determine the mechanical characteristics of such materials is not new [3, 4]. In some studies, the experiments on cyclic loading of the polymer with fixed maximum and minimum of stress (or strain) values in each cycle are described, in other works, the stepwise stretching-compression with increasing amplitude at each loading step is considered [5, 6].

The relevant method was developed to minimize labor expenditures and to obtain in one experiment all necessary data for further theoretical modeling of the elastic-viscous-plastic material behavior. It is based on cyclic uniaxial loading of the sample with increasing strain amplitude. Novelty of the proposed test method consists in entering relaxation stops into the loading cycle when the direction of the motion of gripping clamps changes.

This work presents the experimental and theoretical study of the elastic-viscous-plastic properties of real polyethylene.

1 The experiment

Low density polyethylene grade PE 107-02K ($\rho = 0.91 \text{ g/cm}^3$) was taken as a main research object. Its melting point is 110°C , crystallinity is equal to 40%, and the initial Young's modulus is 73.5 MPa. Mechanical tests were carried out using a tensile testing machine Testometric FS100 and a mechanical sensor FS100kN-CT.

Each cycle of the loading program includes the following operations:

- 1) stretching to some value exceeding the maximum strain obtained in the previous cycle;
- 2) stop of the gripping clamps for a given time for stress relaxation;
- 3) unloading up to the given (but not zero) stress;
- 4) again stop of the gripping clamps for relaxation (the same time period);
- 5) termination of the cycle and beginning of the next cycle (with increasing strain amplitude).

The speed of motion of the gripping clamps under loading–unloading was set 100% per minute. The relaxation time was equal to 10 minutes. The test program consisted of 8 cycles at maximum strain 10%, 20%, 30%, 40%, 60%, 80%, 100%, and 120%. The tensile force during unloading was reduced to 0.6 MPa at each cycle. This regime allowed us to clearly separate the viscoelastic and elastic-plastic parts of the mechanical behavior of the material and to obtain all input parameters necessary for constructing a phenomenological model. As a result, the dependencies of experimental stresses on strain and time (relaxation curves) were obtained.

2 Elastic-viscous-plastic model

The proposed phenomenological model of the elastic-viscous-plastic medium is a further development of the differential approach to the construction of constitutive equations based on the interpretation of the mechanical behavior of the material with the help of symbolic schemes. Previously, this approach has been used to describe the elastic-plastic behavior of polymers. The results of modeling the elastic-plastic properties of nanocomposites based on the polyolefin matrix and ultrafine filler of layered clay minerals (smectites) are presented in [7]. Further, this model has been modernized, which allows us to take into account the changes in the volume of the polymer due to the accumulation of internal damage (micro debondings) [8].

The symbolic model scheme (Fig. 1) consists of two parallel branches containing two serially connected elements: a) elastic (1) and plastic (3), b) elastic (2) and viscous (4).

Tensor equations that specify the properties of the medium correspond to each element.

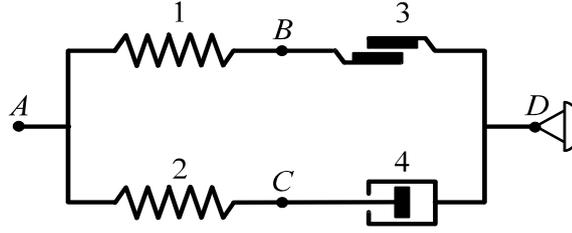


Figure 1: Symbolic scheme of elastic-viscous-plastic model

Stresses in the elements in each branch are the same, and strain rates are summed to a combination of elements. The overall stress in the material is the sum of stresses in the branches, and strain rates in the branches are equal to the total rate of deformation of the material.

The elastic-plastic branch simulates the behavior of agglomerates of tougher crystallites, their displacement, destruction or conversion under deformation. The viscoelastic one describes the flow of the amorphous polymer between the lamellae inside crystallites and in the space around the crystallites and particles.

The equations of the nonlinear elasticity theory are used to calculate Cauchy stress tensors \mathbf{T}_m in elastic elements ($m = 1, 2$). It is assumed that the body volume and temperature in the deformation process do not change, and the elastic properties of the material are described by the neo-Hookean potential:

$$w_m = C_m \left(\text{tr} \mathbf{V}_m^2 - 3 \right),$$

where w_m is the free energy volume density for the m -th element, C_m is the elastic constant, and \mathbf{V}_m is the left stretch tensor, which can be written in terms of a current orthonormal eigenvector triple $\mathbf{n}_i^{(m)}$ ($\lambda_i^{(m)}$ is a corresponding elongation ratio) as

$$\mathbf{V}_m = \sum_{i=1}^3 \lambda_i^{(m)} \mathbf{n}_i^{(m)} \otimes \mathbf{n}_i^{(m)}.$$

True stress tensors for the elastic elements ($m = 1, 2$) of the circuit are determined by formula:

$$\mathbf{T}_m = 2C_m \mathbf{B}_m + p_m \mathbf{I},$$

where \mathbf{I} is the unit tensor, and p_m is the indefinite parameter introduced to take into account medium incompressibility.

The current deformation of elastic elements is determined through the material derivatives of the left Cauchy-Green tensors $\mathbf{B}_m = \mathbf{V}_m \mathbf{V}_m$ ($m = 1, 2$) using evolution equation written for the case of uniaxial strain (no any rotations in the medium):

$$\dot{\mathbf{B}}_m = 2\mathbf{V}_m \mathbf{D}_m \mathbf{V}_m,$$

where \mathbf{D}_m is the strain rate tensor of the m -th elastic element. According to the accepted symbolic scheme these tensors are defined as $\mathbf{D}_1 = \mathbf{D} - \mathbf{D}_3$ and $\mathbf{D}_2 = \mathbf{D} - \mathbf{D}_4$. Tensors \mathbf{D}_3 and \mathbf{D}_4 are calculated using formulas (1) and (2), respectively. The strain rate tensor \mathbf{D} of the entire medium is determined via the deformation gradient of the medium \mathbf{F} :

$$\mathbf{D} = \frac{1}{2} \left[\dot{\mathbf{F}} \mathbf{F}^{-1} + \left(\dot{\mathbf{F}} \mathbf{F}^{-1} \right)^T \right].$$

The mechanical properties of the plastic element are determined by analogy with the basic Prandtl-Reuss equations of plastic flow [9, 10]. To close the system of equations defining the plastic features of the material, proportional relation (1) between the deviators of strain rate tensors of the plastic element \mathbf{D}_3 and the entire medium \mathbf{D} is used:

$$\sqrt{\mathbf{D}_3 \cdot \mathbf{D}_3} = \kappa \sqrt{\text{dev} \mathbf{D} \cdot \text{dev} \mathbf{D}}, \quad (1)$$

where κ is the nonnegative parameter indicating the portion of plastic deformation rate in the total deformation rate of the medium. In the model, this parameter is set through the yield function Φ :

$$\kappa = \begin{cases} 0, & \Phi(\mathbf{V}, \dots) < q, \\ \zeta(q), & \Phi(\mathbf{V}, \dots) = q, \end{cases}$$

where $q = \max \Phi(\mathbf{V})$ is the analog of Odqvist hardening parameter [11] of the classical theory of plasticity (it characterizes the accumulated plastic strain). For the yield function Φ , the intensity of the medium left stretch tensor \mathbf{V} is used:

$$\Phi = \sqrt{\text{dev} \mathbf{V} \cdot \text{dev} \mathbf{V}}.$$

In the case of the incompressible uniaxially loaded material, $\lambda_1 = \lambda$, $\lambda_2 = \lambda_3 = \lambda^{-1/2}$, it can be written as

$$\Phi = \sqrt{2/3} \left(\lambda - \sqrt{1/\lambda} \right).$$

For the viscous element, the stress tensor and its deviator are determined by formula (2) (η is the shear viscosity, $\sigma_0 = 1/3 \text{tr} \mathbf{T}_4$ is the mean normal stress or negative pressure.)

$$\mathbf{T}_4 = 2\eta \mathbf{D}_4 - \sigma_0 \mathbf{I}, \quad \text{dev} \mathbf{T}_4 = 2\eta \text{dev} \mathbf{D}_4 = 2\eta \mathbf{D}_4. \quad (2)$$

Thus, to describe the nonlinear elastic-viscous-plastic behavior of the polymer using this model, we need to know the following four dependencies: $C_1(q)$, $C_2(q)$, $\kappa(q)$ and $\eta(q)$. They were determined from the analysis of relevant experimental data.

The viscous-elastic-plastic properties of polyethylene PE 107-02K were theoretically investigated by this approach. Initial experimental data for modeling were obtained from the mechanical tests of specimens by the method described in the section "Experiment". A comparison of the calculated and experimental results shows that they are practically identical. This is indicative of the fact that the conclusions from the analysis of the findings of model parameters are close to reality. Loading and relaxation curves (true stress versus time or elongation ratio) are shown in Fig. 2.

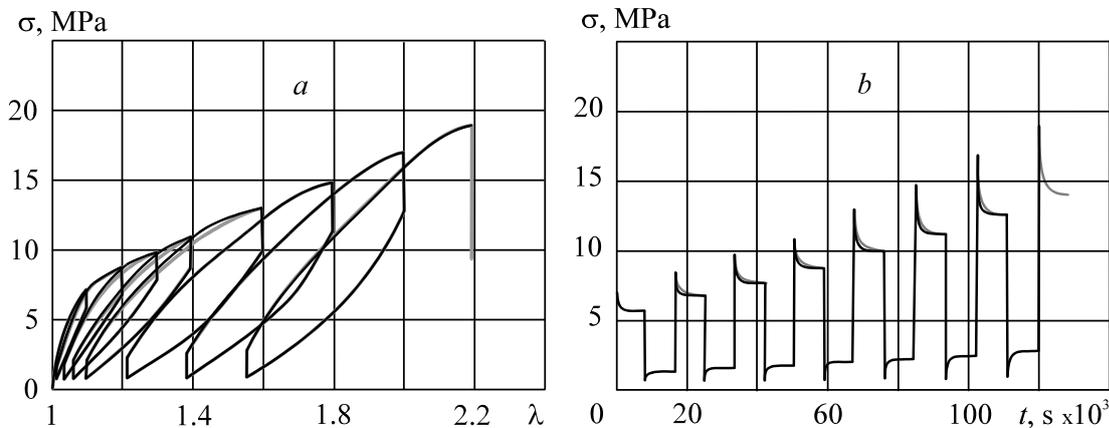


Figure 2: Strain (a) and relaxation (b) cyclic loading curves for PE 107-02K.

Black line — calculation, gray — experiment

The dependences of the model parameters C_1 and κ on q are shown in Fig. 3a and 3b. The choice of such strain measure was done due to the fact that, in contrast to the elongation ratio of the sample λ , the q value cannot decrease during loading. Therefore, the condition of irreversible changes of the above model parameters is put into the model.

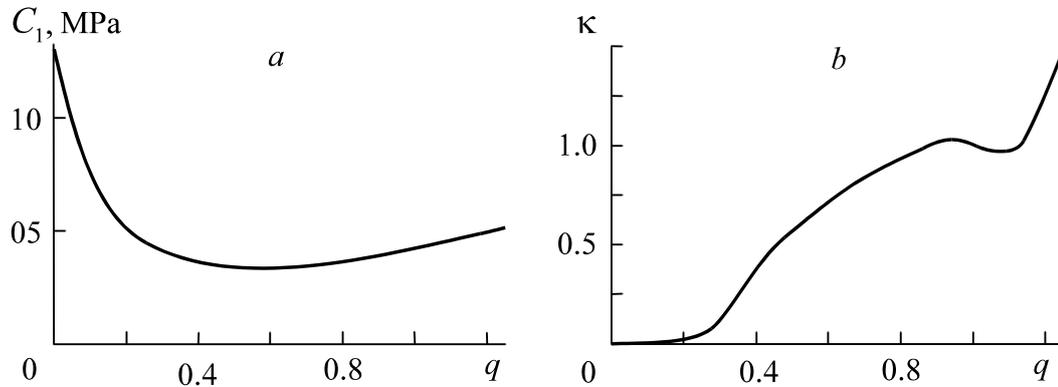


Figure 3: Dependencies of the elastic stiffness C_1 and plasticity parameter κ on strain measure q

The plasticity parameter for PE 107-02K at the elastic stage of deformation (elastic zone corresponds to $q < 0.24$ or $\lambda < 1.2$) is close to zero, then its monotonic increase occurs. The values of κ approach 1 for q equal to about 0.85 ($\lambda = 1.8$), i.e. deformation becomes completely plastic. The curve goes up sharply with a further increase in q ($\lambda > 2$) when the plastic neck formation begins. The plastic neck causes the mechanical inhomogeneity along the sample, but the model does not take it into account. It is supposed that the developed approach will be upgraded in the future using several serially connected four-element schemes to describe this effect.

It has been found that the viscosity η (Fig. 4) increases monotonically over the entire range of loading, i.e., the higher the applied external force, the worse the material flow. The stiffness C_2 that determines the elastic properties of the amorphous phase of the polymer and remains unchanged during deformation is considered constant and taken equal to 10 MPa [12].

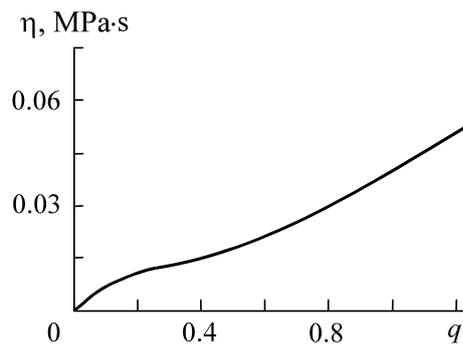


Figure 4: Viscosity η from strain measure q

3 Conclusion

The studies carried out in this work demonstrated the adequacy and validity of the proposed model for investigation of the elastic-viscous-plastic properties of polyolefin polymers. In our opinion, this approach (a combination of new experimental results and the upgraded model) can be successfully applied to obtain useful findings and conclusions not

only about phenomenology, but also about internal processes that occur in the structure of these materials during deformation.

Acknowledgements

This work is executed at a financial support of RFBR Grant 13-08-00065, Perm Ministry of Industry of Innovations and Science (Grant 14-08-96013 p_ural_a) and Program RAS 12-C-1-1015.

References

- [1] Fischer J.M. Handbook of Molded Part Shrinkage and Warpage. Plastics Design Library/William Andrew Inc., 2003. 263 p.
- [2] White J.L., Choi D. Polyolefins: Processing, Structure Development and Properties. Hanser Publishers, 2005. 271 p.
- [3] Meyer R.W. Pruitt L.A. The effect of cyclic true strain on the morphology, structure, and relaxation behavior of ultra high molecular weight polyethylene. *Polymer*. 2001. V. 42. P. 5293–5306.
- [4] Bergstrom J.S., Rimnac C.M., Kurtz S.M. An augmented hybrid constitutive model for simulation of unloading and cyclic loading behavior of conventional and highly crosslinked UHMWPE. *Biomaterials*. 2004. V. 25. P. 2171–2178.
- [5] Ayoub G., Zairi F., Nait-Abdelaziz M., Gloaguen J.M. Modelling large deformation behaviour under loading-unloading of semicrystalline polymers: application to a high density polyethylene // *Int. J. Plasticity*. 2010. V. 6. P. 329–347.
- [6] Drozdov A.D., Klitkou R., Christiansen J.C. Cyclic viscoplasticity of semicrystalline polymers with finite deformations // *Mechanics of Materials*. 2013. V. 56. P. 53–64.
- [7] Garishin O.K., Svistkov A.L., Gerasin V.A., Guseva M.A. Simulation of the elastic-plastic behavior of polyolefin-based nanocomposites with a different structure of layered filler // *Polymer Science. Ser. A*. 2009. V. 51, No 4. P. 407–415.
- [8] Garishin O.K., Gerasin V.A., Guseva M.A. A study of the elastoplastic properties of polymer-silicate nanocomposites with allowance for the change in their volumes during deformation. *Polymer Science. Ser. A*. 2011. V. 53, No 12. P. 1187–1197.
- [9] Prandtl L. Spannungsverteilung in plastischen Körpern. *Proc. I. Int. Congress Applied Mech. Delft*. 1924. S. 43–4.
- [10] Reuss A. Berücksichtigung der elastischen Formänderung in der Plastizitätstheorie. *Ztschr. Angew. Math. Und Mech*. 1930. Bd. 10, H. 2. S. 266–274.
- [11] Odkvist F.K.G. *Mathematical Theory of Creep and Creep Rupture*. Stockholm: Darendon Press, 1966. 170 p.
- [12] Oleinik E.F. Plasticity of semicrystalline flexible-chain polymers at the microscopic and mesoscopic levels. *Polym. Sci. Ser. C*. 2003. V. 45, No 1. P. 17–117.

Investigation of the elastic-viscous-plastic behavior of thermoplastic materials.
Experiment and simulation

Oleg K. Garishin, Institute of Continuous Media Mechanics UB RAS, Perm, Russia

Anton S. Korlyakov, Institute of Continuous Media Mechanics UB RAS, Perm, Russia

Vladimir V. Shadrin, Institute of Continuous Media Mechanics UB RAS, Perm, Russia