Graded Orientation of the Linear Polymers

Levan Nadareishvili, Roland Bakuradze, Barbara Kilosanidze, Nona Topuridze, Liana Sharashidze, Ineza Pavlenishvili

***Abstract*—**Some regularities of formation of a new structural state of the thermoplastic polymers - gradually oriented (stretched) state (GOS) are discussed. Transition into GOS is realized by the graded oriented stretching - by action of inhomogeneous mechanical field on the isotropic linear polymers or by zone stretching that is implemented on a standard tensile-testing machine with using a specially designed zone stretching device (ZSD). Both technical approaches (especially zone stretching method) allows to manage the such quantitative parameters of gradually oriented polymers as a range of change in relative elongation/orientation degree, length of this change and profile (linear, hyperbolic, parabolic, logarithmic, etc.). The possibility of obtaining functionally graded materials (FGMs) by graded orientation method is briefly discussed. Uniaxial graded stretching method should be considered as an effective technological solution to create polymer materials with a predetermined gradient of physical properties.

***Keywords*—**Controlled graded stretching, gradually oriented state, linear polymers, zone stretching device.

I. Introduction

UNIAXIAL oriented stretching is a widespread method of the structural modification of the linear polymers [1]-[3]. As a result of stretching at the above glass-transition temperature, the isotropic polymer goes over into the oriented state, which is characterized by a preferential location of the structural elements in the stretching direction. Polymer becomes anisotropic with symmetry similar to symmetry of an uniaxial crystal. The structural anisotropy leads to considerable anisotropy of such physical properties as optical, electrical, acoustic, thermal, mechanical, sorption, etc. The samples with different physical and mechanical properties may be obtained by means of the variation of the orientation degree depending on the stretching temperature and rate, a relative elongation $\Delta l/l$ (Δl is a real elongation, l is the initial length of the sample) and the cooling rate. In the samples oriented in the conventional mode the relative elongation is practically the same and, accordingly, the anisotropy is the same.

Uniaxial oriented stretching can be carried out in a different mode. Earlier we have established the conception about new structural state of the thermoplastic polymers – gradually oriented (stretched) state (GOS) presented in [4]–[11] and offered the appropriate technical solution for uniaxial graded oriented stretching for transition of isotropic polymers into GOS [12]–[14].

According to this conception, as a result of the transformation of isotropic polymers and their composites to GOS, the gradient of all the properties (optical, electrical, acoustic, thermal, mechanical, sorption, etc.) should be generated in materials that depend on the value of relative elongation/orientation degree.

The logic is simple: as the structural anisotropy, arising as a result of the uniaxial stretching, leads to anisotropy of physical properties, it should be apparent that the gradient anisotropy, arising as a result of uniaxial graded stretching should lead to gradient of physical properties. Let us show the validity of these considerations on the example of the birefringence.

The birefringence resulting from uniaxial tension is a function of the relative elongation

$$\Delta n = n_1 - n_2 = \gamma\lambda \tag{1}$$

where n_1 and n_2 are the refraction indices of the ordinary and extraordinary rays, respectively; γ is optical deformation coefficient; λ is the relative elongation ($\Delta l/l$). It follows from this equation that gradient of the relative elongation should cause the gradient of birefringence.

Based on these, we can conclude that the graded stretching method provides the wide possibilities for creating new type materials/elements with the gradient of various physical properties. The isotropic polymer transition into GOS may be realized by the action of inhomogeneous mechanical field on the sample as well as by zone stretching that is implemented by displacement the heating zone and tensile speed changing in the stretching process. Designing options for inhomogeneous mechanical field are unlimited. Consequently it is not feasible to develop a unified, comprehensive technical solution of graded stretching process. Therefore the development of each new graded oriented stretching method is an important part in the study of scientific and technological aspects of the GOS and the various perspective applications.

This article attempts to demonstrate the fruitfulness of the concept of GOS. For this purpose the controlled uniaxial graded oriented stretching is considered for some specific configurations of inhomogeneous mechanical field. Zonal stretching method is also discussed which is carried out on a standard tensile-testing machine with the use a specially designed zone stretching device (ZSD). The possibility of obtaining functionally graded materials (FGMs) by graded orientation method is briefly discussed.

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II. Controlled Graded Oriented Stretching Methods

A. Stretching in Inhomogeneous Mechanical Field (Graded Stretching of Trapezoidal Samples)

Let's consider rectangular Cartesian co-ordinates XOY, where the abscissa is parallel to the large base of trapezoid ABCD and the ordinate is an axis of symmetry (Fig. 1).

Introduce the notations: $AD \equiv 2a$, $BC \equiv 2b$, $NM \equiv H$, $NP_1 \equiv h$. $P_1P_2 \equiv l$, the elongation of P_1P_2 is a segment $P_2P_3 \equiv \Delta l$.

Let's assume that the sides of trapezoid are $y = f(x)$, $x \in (0, a]$ function. MN is an axis of symmetry. $P_1 \in MN$. Then we get:

$$h = f(l) \quad (2)$$

$$l = f^{-1}(h) \quad (3)$$

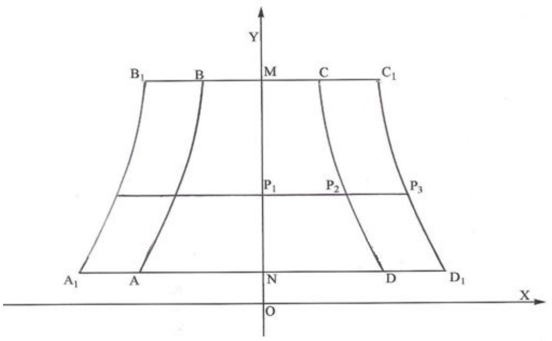


Fig. 1 Scheme of graded stretching of trapezoidal sample. Explanations are given in the text

During stretching the sides of the trapezoid moved parallel to its initial position at a distance Δl. After stretching the sample takes a form of curvilinear $A_1B_1C_1D_1$–trapezoid. $P_3 \in C_1D_1$. In this case the elongation of the segment is $P_2P_3 \equiv \Delta l$ = const.

The quantitative parameters of the gradually oriented sample $A_1B_1C_1D_1$ are: the changing range of the relative elongation $\frac{\Delta l}{ND} \div \frac{\Delta l}{MC}$; changing length of the relative elongation is *MN*.

Distribution of relative elongation in height of the trapezoid is expressed by the equation:

$$\frac{\Delta l}{l} = \frac{\Delta l}{f^{-1}(h)} \quad (4)$$

Let's consider the various cases of function:

a) In the case of linear function $f(x) = kx + c \;\Rightarrow\; f^{-1}(h) = \frac{h-c}{k}$, we get

$$\frac{\Delta l}{l} = \frac{k \cdot \Delta l}{h - c}$$

After introduction of the notation $m = \Delta l \cdot k$, we get

$$\frac{\Delta l}{l} = \frac{m}{h - c} \quad (5)$$

b) In the case of quadratic function $f(x) = x^2 \;\Rightarrow\; f^{-1}(h) = h^{\frac{1}{2}}$, we have

$$\frac{\Delta l}{l} = \Delta l \cdot h^{-\frac{1}{2}} \quad (6)$$

c) The logarithmic function $f(x) = \ln x \;\Rightarrow\; f^{-1}(h) = e^h$ gives

$$\frac{\Delta l}{l} = \Delta l \cdot e^{-h} \quad (7)$$

d) In the case of hyperbolic function $f(x) = \frac{k}{x} \;\Rightarrow\; f^{-1}(h) = \frac{k}{h}$, we get

$$\frac{\Delta l}{l} = \frac{\Delta l \cdot h}{k}$$

Introduce the notation $m = \frac{\Delta l}{k}$, then we get

$$\frac{\Delta l}{l} = m \cdot h \quad (8)$$

Similar approach can also be applied to other functions. In the case, when $f(x)$ or $\frac{\Delta l}{l}(h)$ are complex functions the quantitative technique can be applied.

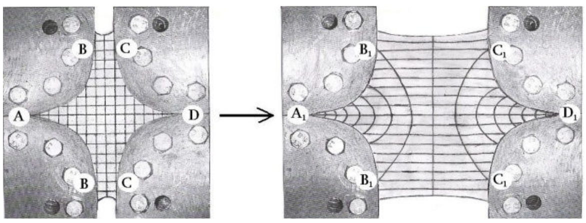
For the experimental confirmation of the presented theoretical data we have considered the samples of two types having a trapezoid shape with hyperbolic and linear sides.

Fig. 2 (a) shows the isotropic polyvinyl alcohol (PVAL) film, which consists of two isosceles trapezoids *ABCD* with hyperbolic ($y = K/x$) sides, having one shared large *AD*–base. The use of the dual trapezoid excludes narrowing of the sample during stretching from large *AD*–base. The following trapezoid-sample parameters were used: the small base *BC* = 10 mm, the large one *AD* = 80 mm, the height *h* = 35 mm, the thickness was 0,2 mm, *K* = *xy* = 156. The films were stretched on the strength testing machine at 358^0K with rate of 1,5 mm/min by 100 - 500% with respect to the free side of the film (i.e. to small *BC*–base of the trapezoid). The overlay square grid was applied to register the elongation distribution of the film. Fig. 2 (b) shows the same PVAL–film stretched by 500%. The topographic pattern clearly indicates to the existence of the elongation gradient.

The relative elongation distribution in the gradually stretched PVAL–films was calculated.

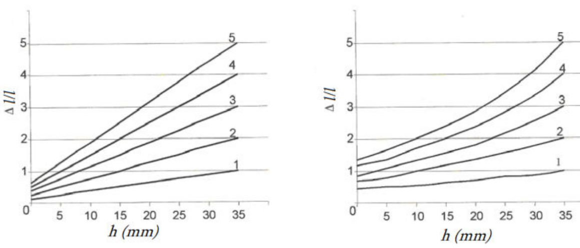
Fig. 3 (a) shows the relative elongations distribution in the films with hyperbolic sides obtained theoretically from (8), which is derived at the supposition that the elongation distribution is uniform in the stretching direction.

But Fig. 2 (b) shows that this supposition is not strictly complied. The elongation distribution in the stretching direction is inhomogeneous; the middle part of the sample is deformed to a greater degree, especially closer to the large A_1D_1 –base of the trapezoid. In this case the actual distribution of elongations can be described by assemblage of appropriate curves. We confine our discussion to such a part of the films, which is formed by graded stretching along the small BC–base of trapezoid. In Fig. 3 (b) the experimental results are given. A comparison of theoretical and experimental data shows a certain differences, in particular, nonlinear distribution of the relative elongation in real samples. This nonlinearity increases with the degree of stretching. Besides, the range of changing of the relative elongation is less in real samples. For example, the range of changing is of 0.625 – 5 in $\Delta l/l$ at 500% stretch (in theory) and of 1.35 – 5 (actually).



(a) (b)

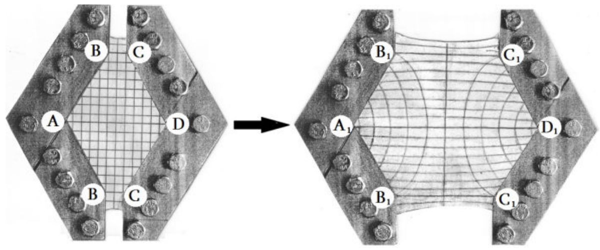
Fig. 2 Graded stretching of PVAL trapezoidal film. (a) isotropic film (two trapezoids *ABCD* having hyperbolic form sides with one shared large *AD*–base) attached between clamps of stretching device; (b) anisotropic film (two trapezoids $A_1B_1C_1D_1$ having hyperbolic sides with one shared large A_1D_1 –base) formed by stretching of the isotropic film by 500% (calculated with respect to a small *BC*–base). The stretching direction is indicated by the arrow



(a) (b)

Fig. 3 Dependence of the relative elongation $(\Delta l/l)$ on the height (*h*) for $A_1B_1C_1D_1$ –trapezoidal in PVAL–films with hyperbolic sides stretched on: 1 – 100%; 2 – 200%; 3 – 300%; 4 – 400%; 5 – 500%. (a) theoretical calculations; (b) experimental data

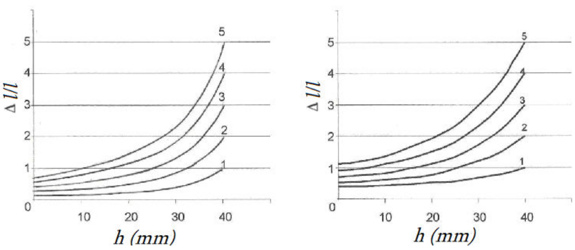
Fig. 4 (a) shows the isotropic PVAL–film consisting of two isosceles *ABCD*–trapezoids with linear sides, having one shared large *AD*–base. The trapezoid-sample parameters were: the small base *BC* = 10 mm, the large one *AD* = 60 mm, the height *h* = 40 mm, the thickness is 0,2 mm. The films were stretched at 358^0K and at the rate of 1,5 mm/min on 100–500% with respect to the free side of the film (to small *BC*–base of the trapezoid). Fig. 4 (b) shows the same PVAL–film stretched by 500%. The topographic pattern clearly indicates also to the existence of the gradient of elongation. In contrast to the previous one, in this case, visually the elongation distribution is almost equal in the stretching direction.



(a) (b)

Fig. 4 Graded stretching of PVAL trapezoidal film. (a) isotropic film (two trapezoids *ABCD* having linear sides with one shared large *AD*–base) attached between the clamps of a stretching device; (b) anisotropic film (two trapezoids having linear sides with one shared large A_1D_1 –base) formed by stretching of isotropic film by 500 % (calculated with respect to a small *BC*–base). The stretching direction is indicated by an arrow

The relative elongation distribution in the gradually stretched PVAL-film was calculated.



(a) (b)

Fig. 5 Dependence of the relative elongation $(\Delta l/l)$ on the height (*h*) of $A_1B_1C_1D_1$ –trapezoidal film with linear sides stretched on: 1 – 100%; 2 – 200%; 3 – 300%; 4 – 400%; 5 – 500%: (a) theoretical calculations; (b) experimental data

Fig. 5 (a) shows the theoretically obtained distribution of the relative elongations in the films with linear sides calculated according to (5) which are derived under supposition that the elongation distribution is uniform in the stretching direction.

Fig. 5 (b) shows experimental curves calculated for such a part of the film, which is formed by graded stretching of the area along the small BC- base of trapezoid (as well as in Fig. 3 (b)).

It is important to note that a rather good correspondence between the theoretical calculations and experiment is established. Some differences can be eliminated by correction of the mechanical field configuration (geometry of the sample/profile of clamps).

B. Stretching in Inhomogeneous Mechanical Field (Graded Stretching of the Curvilinear Trapezoidal Sample)

In the above examples the lower range of changes in the relative elongation is always more than zero and the larger, the higher is the value of the deformation.

It is therefore of interest to develop such an algorithm of gradient orientation, when the lower limit of the range of changes in relative elongation would be zero. The method for obtaining the rectangular samples with the following parameters is suggested below:

- range of change in relative elongation $\Delta l / l$ is 0 ÷ n;
- length of change in relative elongation is *h*;
- profile of the relative elongation distribution is hyperbola.

Such samples can be obtained by uniaxial stretching of the curvilinear trapezoidal *ABCDM* film (Fig. 6). The arrow indicates the direction of stretching.

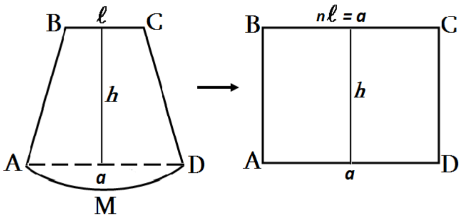


Fig. 6 Scheme of transition of the curvilinear trapezoidal film to rectangular one. Explanations are given in the text

Let us introduce the following notations:

- length of arc–*AMD* is denoted by *a*;
- a small *BC*-base of the trapezoid – through *l*;
- the distance between the segments *AD* and *BC* – through *h*;
- the multiple relative deformation of *BC*–base – through *n*.

It is required to compute the geometric dimensions of the *ABCDM* –trapezoid. The values of *l, h* and *n* are given. The task is in determination of the *R*–radius of the arc–*AMD* and its degree measure β when the condition $a = nl$ is fulfilled.

After simple transformation we obtain the following equations:

$$0{,}017\beta\left(\frac{2m}{\cos\beta}+\frac{1}{\sin\beta}\right)=n \tag{9}$$

$$R=\frac{180\cdot na}{\pi\beta} \tag{10}$$

where *m* = *h/l*. To calculate β from (9) an algorithm based on the method of chords and the appropriate program in the algorithmic language PASCAL were prepared. Further, radius *R* is determined from (10), and the sample is made for experiment.

Fig. 7 illustrates the graded stretching technique. The curvilinear *ABCDM*–trapezoid with computed sizes is fixed between the clamps of the stretching device disposed mutually parallel. The distance between the clamps equals to the length *l* of a small base of trapezoid. The trapezoidal test sample is fixed in the clamps along the sides *AB* and *CD*. If we move the clamp (the shift direction is indicated by an arrow), the stretching front moves smoothly from a small *b*–base of the trapezoid to the arc–*AMD*. Stretching of the sample was stopped when $l+\Delta l$ (Δl increase in the length *l* of a small base of the trapezoid) will be equaled to the length of arc–*AMD*, which becomes a straight line at this point.

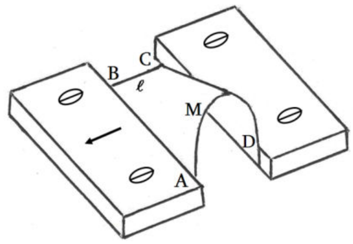
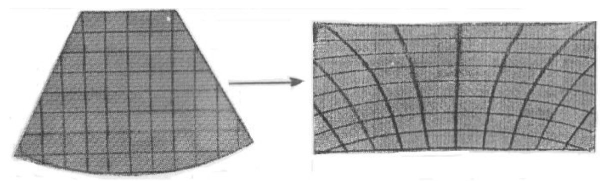


Fig. 7 Illustration of the graded stretching technique. Explanations are given in the text

As a result of stretching the curvilinear trapezoid transforms to a rectangle with sides *b* and *h*. In the described mode of graded stretching the lower limit of change in elongation is almost zero and the upper limit is determined by the ratio *a /l.*

Experiments were carried out with isotropic PVAL–films (the thickness 0.2 mm, the stretching temperature 358^0(K)). In order to visualize the distribution of elongations on the original sample the square grid was applied. Fig. 8 (a) shows the original curvilinear trapezoid-sample for which *a/l* = 3 (*a* = 60 mm, *l* = 20 mm) and *h* = 30 mm. After a three-fold extension (relatively to a small base of the trapezoid) the sample is transformed into a rectangle (Fig. 8 (b)). Changing in the topographic pattern of a square grid clearly indicates the existence of the gradient elongation perpendicularly to the stretching direction.



(a) (b)

Fig. 8 Transition of curvilinear trapezoidal isotropic film (a) into rectangular gradually oriented one (b)

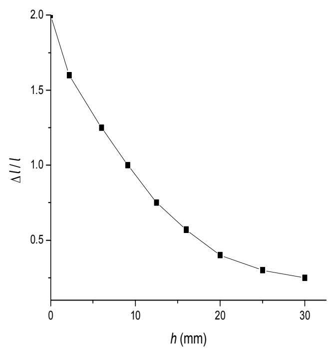


Fig. 9 Relative elongations distribution in the central part of the gradually oriented rectangular film (Fig. 8 (b))

Fig. 9 shows the relative elongations distribution in the central part of the gradually oriented rectangular film (Fig. 8 (b)).

According to the Fig. 9 we get:

- the range of change in relative elongation is 0.225 ÷ 2;
- the length of change in relative elongation is *h* = 30mm;

- the profile of the relative elongation distribution is hyperbola.

We believe that the experimental data agree quite well with theoretical calculations.

III. Zone Graded Stretching

Zone stretching can be carried out on a standard tensile-testing machine with using the zone stretching device (ZSD), proposed in [15].

The device contains a heater which is disposed between the fixed and active clamps of the tensile-testing machine. Between the heater and an active clamp the cooler is placed in the form of a blower, a sylphon bellows with liquid or the cylindrical tubes rotating around their axis and in which the liquid circulates. A heater and air blower have preferably equidistant geometric shape with respect to the cross section of the test sample. The heater is done with the ability of heating the sample by the width of the selected area with the selected size and profile, and the cooler is done with the ability to direct on this area a stream of air, to cover liquid from sylphon bellows or rolling the cylinders rotating around their axis on the surface of the test film.

Zone stretching is carried out in the following way: we placed a heater and a cooler near the active clamp. When the selected temperature is achieved, the simultaneous movement of all three mobile units of the device (the active clamp, a heater and a cooler) begins. In this case a heater and a cooler are moved in a direction opposite to the movement of the active clamp. In the heating zone, the value of the yield strength of the polymer is minimum. It is why only the heated section of the sample is stretched. As a result of stretching and displacement of the heater this area of the sample goes out of the heating zone and enters into the cooling zone. As a result of the simultaneous movement of three mobile units of the device, the stretching process is gradually spreading towards the movement of the heater and cooler. The stretching mode (temperature and speed) can be changed at any stage.

ZSD makes it possible to implement a very wide range of stretching modes, in particular:

- localization of the stretching process in the frames of selected zone of the sample;
- the possibility of creating different front of stretching;
- stretching of sample in the continuous and/or jumping regime with constant rate and/or acceleration;
- conducting of stretching in homogeneous or gradient regime;
- realization of uniaxial stretching of different selected region of one and the same sample in the different regime (temperature, rate and value of deformation) at temperature higher than room temperature without taking out of the sample from clips;
- with use of the equipment for stretching of polymer films at temperature higher than room temperature in accordance with above item it is possible to define the influence of deformation rate and temperature on such mechanical parameters of separate regions of the sample, investigation of which do not requires the destruction of whole sample.

These characteristics are:

- mechanical stress and deformation according to the Hooke's region;
- modulus of elasticity;
- flow modulus;
- mechanical stress at the fixed deformation;
- relaxation of mechanical stresses.

The stretch degree of the sample is determined by the speed ratio of the displacement of active clamp and a heater. The gradient stretching mode is achieved by varying this ratio in the sample stretching process. Preselected experiment mode is carried out via the control unit. A more complete description of the ZSD is given in [15].

Some illustrative materials are presented below.

Fig. 10 shows the gradually oriented rectangular PVAL–films (the thickness 0.2 mm) stretched at 358^0K by using ZSD.

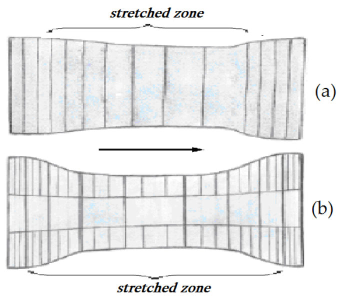


Fig. 10 Gradually stretched rectangular PVAL–films. The overlay parallel lines are applied to register the elongation distribution of the film. The arrow indicates the direction of propagation of the stretch front

Fig. 11 shows the distribution of relative elongations along the length of the same gradually oriented films.

In Fig. 11 (a) the relative elongation over the length of the film varies linearly within 0 ÷ 230% in case of a total lengthening of 60%. In Fig. 11 (b) the distribution of relative elongation first increases and then decreases along the length of the film. In case of a total film lengthening of 85% the relative elongation varies over the length within 0 ÷ 250% ÷ 0.

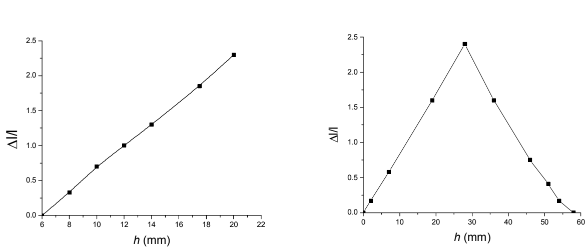


Fig. 11 Relative elongations distribution in the gradually oriented rectangular film: (a) Fig. 10 (a); (b) Fig. 10 (b)

Conception about a new structural state of the linear polymers – about GOS, the formulation of GOS's quantitative parameters, the development of methods of obtaining the gradually oriented polymers with the pre-determined parameters (as well as methods that will undoubtedly be developed in the future) appreciably expand the problems of the study of polymer science. The uniaxial graded oriented stretching method can be considered as scientific and technological innovation for the creation of a new type of FGMs/elements on the base of linear polymers/copolymers and its composites. The creation and investigation of FGMs is one of the strategic directions in modern materials science [16]–[18]. FGMs are composites or single-phase materials, whose functional properties change uniformly or abruptly at least in one dimension of the particle, film or a bulk sample. A change in the properties of FGMs is associated with a corresponding variation of the chemical composition or physical structure of the material.

Graded oriented stretching method gives the possibility to create the FGMs using both known methods – creation of gradient of microstructure or gradient of chemical composition.

In our case gradient of microstructure corresponds to the gradient of preferential location of the structural elements – macromolecular chains and its sections in the stretching direction which is characterized quantitatively by the gradient of orientation/stretching degree. Gradient of microstructure causes the gradient of optical, mechanical, thermal, acoustic, sorption, etc. properties of polymer material. Gradient of microstructure can be formed on the base of single polymer/copolymer. For illustration we can call GB (Gradient Birefringence) – elements, which were obtained by gradient orientation of PVAL and PET–films [4]–[8].

Gradient of chemical composition is created by stretching of polymer composites (in which thermoplastic polymer is a matrix for metal, ceramics, another polymer or pores) in graded mode. For example, in polymer composites, containing conductive particles the gradient orientation leads to a gradient redistribution of the concentration of conductive particles in the stretching direction and consequently to the gradient of electrical resistance. The same reasoning is also true for magnetic composites. Our experiments, conducted on PVAL–composites contained conductive particles (graphite, carbon black) and magnetic particles (Ni, Fe_3O_4), confirm this argument [19].

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