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APPLICATION OF THE FRACTAL APPROACH TO DETERMINATION OF THE POISSON COEFFICIENT OF POLYMERIC SYSTEMS

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Within the framework of the combination of the classical elasticity theory with the concepts of the scale invariance of the polymer structure, the dependence of the Poisson coefficient on the dimension of the fractal cluster has been found. It has been shown that the Poisson coefficient of polymers is related to the value of relative deformation. The range of permissible values of the fractal dimension of materials for the cases of uniaxial tension and compression has been determined.

In theoretical approaches to description of the deformed state of polymeric systems, use is generally made of different model representations of the substance structure which are practical in the form of additional limiting conditions. These limitations result in reducing the number of undetermined deformation parameters to a minimum. This in turn does not allow one to account in full measure for a change in the structural factors of polymers, which dominate in formation of the corresponding properties of the system. These factors also include the geometry of deformed structure formations.

According to the classical theory of entropy elasticity, the deformation behavior of polymers in a highly elastic state is described by an equation of the form [1]

$$W = A\,(\lambda_1^2 + \lambda_2^2 + \lambda_3^2 - 3)\,, \tag{1}$$

where λ_i (i = 1, 2, 3) are the multiplicities of deformation along three mutually perpendicular axes. In the case of uniaxial deformation (for example, along the axis $i = 1$), expression (1) is reduced to a one-parameter equation due to the superposition of additional conditions. In particular, in an isotropic material, account for the incompressibility condition $\lambda_1\lambda_2\lambda_3 = 1$ leads to the expression

$$\lambda_2 = \lambda_3 = \lambda_1^{-1/2} \tag{2}$$

and respectively to

$$W = A\,(\lambda_1^2 + 2\lambda_1^{-1} - 3)\,.$$

We note that condition (2) automatically excludes from consideration materials with a negative Poisson coefficient, whose existence has been confirmed experimentally among both polymers [2] and crystals of cubic and hexagonal structures [3, 4]. For this reason, the relation

$$\varepsilon_{\perp} = -\,\nu\varepsilon_{\parallel}\,, \tag{3}$$

which determines the development of lateral deformations in the absence of the corresponding external actions [5], has a great physical content, even though it leads to the appearance of a structural parameter ν in the expression for a highly elastic potential:

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$$W = A\,[\lambda_1^2\,(1 + 2\nu^2) - 4\nu\,(\nu + 1)\,\lambda_1 + 2\nu^2 + 4\nu - 1]\,. \tag{4}$$

In [6], based on the experimental data on equilibrium uniaxial tension, it was shown that in many cases the deformation behavior of cross-linked rubbery polymers is described more exactly by the Bartenev–Khazanovich highly elastic potential

$$W = B\,(\lambda_1 + \lambda_2 + \lambda_3 - 3)\,. \tag{5}$$

Without going into detailed analysis of the conditions for applying relations (1) and (5), we can note that their formal difference is initiated by the features of the structural organization of the systems under study, which are manifested at the spatial-topological level. Indeed, the results of investigations carried out in [7], as applied to self-similar fractal structures, make it possible to obtain a general expression for the highly elastic potential in the form

$$W = C\left(\sum_{i=1}^{d} \lambda_i^{d_\mathrm{f}} - d\right), \quad C = \mathrm{const}\,. \tag{6}$$

Therefore, following the concepts accepted in fractal elasticity theory, it can be ascertained that Eqs. (1) and (3) describe the deformation behavior of polymers with different conformation characteristics of macromolecules.

One question that we will consider next can be formulated as follows: to what extent does the classical determination of the Poisson coefficient (3) agree with scaling relations describing the self-similarity of elastic-isotropic fractals. This formulation with certain general limitations is equivalent to the elucidation of the degree of independence of the fundamental propositions postulated accordingly in the classical and fractal elasticity theories.

Let us assume that l_i and L_i are the characteristic dimensions of the fractal in the d-dimensional space before and after deformation. Here, the volumes bounding the fractal in the initial and final states are expressed in the following form:

$$V_0 = \prod_{i=1}^{d} l_i\,, \quad V = \prod_{i=1}^{d} L_i\,.$$

The self-similarity of the elastic-isotropic fractal is retained provided that the change in the volume due to the deformation obeys the scale-invariant relation [8]

$$V = V_0\,\lambda_\mathrm{f}^{\alpha}\,, \tag{7}$$

where $\alpha = d - d_\mathrm{f}$ and λ_f is the change in the characteristic scale L_f due to the elastic deformation. If the deformation is uniaxial ($\lambda_\mathrm{f} = \lambda_1$) and, by virtue of the isotropy, all the transverse changes in the scales are equal to each other ($\lambda_\perp = \lambda_j$, $j \neq 1$), then

$$\frac{V}{V_0} = \prod_{i=1}^{d} \lambda_i = \lambda_1 \lambda_\perp^{d-1}\,,$$

whence, with account for relation (7), we obtain

$$\lambda_\perp = \lambda_1^{1 - \frac{d_\mathrm{f}}{d-1}}\,. \tag{8}$$

Within the framework of the fractal approach it has been shown [8] that under the deformation conditions described the requirement for retaining the self-similarity d_f = const is equivalent to the dependence

$$\lambda_{\perp} = \lambda_1^{-\nu} , \tag{9}$$

where the Poisson coefficient is introduced as the analog of the critical index in the theory of phase transitions of the second kind [9].

From comparison of expressions (8) and (9) we obtain the dependence of the Poisson coefficient of the elastic-isotropic fractal on its dimension d_{f} and the dimension of the bounding space d:

$$\nu = \frac{d_{\mathrm{f}}}{d - 1} - 1 . \tag{10}$$

It is characteristic that it is possible to obtain the same result proceeding from the classical expression (3) in the case of superposition of additional conditions. Indeed, in the general solution of Eqs. (3) and (8) for λ_1, we have

$$1 - \nu (\lambda_1 - 1) = \lambda_1^{\gamma} ,$$

where $\gamma = 1 - \frac{d_{\mathrm{f}}}{d - 1}$. Here

$$\gamma = \frac{\ln [1 - \nu (\lambda_1 - 1)]}{\ln \lambda_1} .$$

Expanding the numerator and the denominator in a series

$$\ln [1 - \nu (\lambda_1 - 1)] = - \nu (\lambda_1 - 1) - \frac{1}{2} \nu^2 (\lambda_1 - 1)^2 + \ldots , \quad \ln \lambda_1 = \lambda_1 - 1 - \frac{1}{2} (\lambda_1 - 1)^2 + \ldots ,$$

and confining ourselves to the first terms of the expansion, we obtain

$$\gamma \approx \frac{- \nu (\lambda_1 - 1)}{\lambda_1 - 1} = - \nu \tag{11}$$

or

$$\nu = \frac{d_{\mathrm{f}}}{d - 1} - 1 . \tag{12}$$

Thus, the fractal and the classical first-approximation approaches give identical analytical results. It should be noted that relation (11) is satisfied for $0 < \lambda_1 \neq 1 \leq 2$ (the interval of possible values of λ_1 is determined by the conditions of expansion of the logarithmic function in a series); for all the possible values of the Poisson coefficient we have $-1 \leq \nu \leq 1/2$. In conformity with this, the values of the fractal dimension of the deformed medium in the accepted system of approximations are limited by the interval

$$d - 1 \leq d_{\mathrm{f}} \leq \frac{3}{2} (d - 1) . \tag{13}$$

However, this result contradicts Eq. (5), wherein the dimension of the bounding space $d = 3$ corresponds to $d_{\mathrm{f}} = 1$. In this connection, some remarks need to be made on the essence of this question.

As we believe, the conclusion, following from relation (10), that the Poisson coefficient is uniquely determined only by the spatial-topological characteristics reveals the physical essence of the problem somewhat unilaterally. The deformation characteristics of polymers depend on different (including also structural) factors: the degree of crystallinity, flexibility, and branching of macromolecular chains as well as on the level of supermolecular organization [10–13]. This, in particular, is also true in full measure of the Poisson coefficient. Therefore, even materials possessing a chemical structure of the same type exhibit the dependence of the quantity ν on the parameters which are determining in formation of a set of their properties. In [14], using polyethylene as an example, it has been shown that different values of the Poisson coefficient correspond to dissimilar degrees of macromolecular branching. The dependence of ν on the value of the relative deformation has also been analyzed in [14]. In addition, it has been shown that in the region of deformations up to 60%, the values of the Poisson coefficient grow with ε, during which the difference in the quantities $d\nu/d\varepsilon$ is observed for materials with dissimilar conformation characteristics.

Thus, it should be expected that the Poisson coefficient depends on the scale factor λ_1 (or on the relative deformation $\varepsilon_{\|}$). This dependence can be taken into account in practice on examining the subsequent terms of expansion of logarithmic functions. In this case,

$$1-\frac{d_{\mathrm{f}}}{d-1}=-\frac{2\nu+\nu^2(\lambda_1-1)}{2-(\lambda_1-1)},$$

whence

$$\nu=-k\pm\sqrt{k^2+\left(\frac{d_{\mathrm{f}}}{d-1}-1\right)(2k-1)}\;, \tag{14}$$

where $k = 1/(\lambda_1 - 1) = \varepsilon_{\|}^{-1}$. We note that the signs before the radical in Eq. (13) are determined only by the direction of deformation: the minus sign corresponds to the uniaxial compression $(0 < \lambda_1 < 1)$ of the elastic-isotropic fractal, while the plus sign corresponds to the uniaxial tension $(1 < \lambda_1 \le 2)$.

The result obtained shows that although the set of properties of the actual material "is coded" in its structure, which in turn is determined by the method of physical condensation, the specific numerical values of the physical parameters are an adequate reflection of the action of external fields of different nature on the system. Therefore, only in the first approximation can the Poisson coefficient be considered to be a constant of a given material, since the Poisson coefficient, being a structurally sensitive characteristic of the material, must depend on the stressed state formed by the external action. Moreover, the fractality of the material structure is manifested beginning with an isolated minimum scale which, in the case of the deformation process, depends on the intensity of the load applied [15]. Figuratively speaking, at each intermediate stage of deformation, the quantity ν acquires a local "induced" value, whereas the averaging has meaning only on such deformation scales to which there corresponds a constant value of the fractal dimension.

It is characteristic that in the case of uniaxial tension the range of permissible values of the fractal dimension is limited to the interval

$$1 \le d_{\mathrm{f}} \le \frac{3}{2}(d-1)\,, \tag{15}$$

whereas for uniaxial compression this interval is somewhat narrower:

$$1 \le d_{\mathrm{f}} \le \frac{5}{4}(d-1)\,. \tag{16}$$

In any case, these intervals are much wider than their analog (13) obtained in the first approximation. In particular, the lower limit of this interval $d_{\mathrm{f}}^{\mathrm{min}} = 1$ is independent of the dimension of the bounding space and corresponds to the exponent of the deformation multiplicities in relation (5).

As follows from Eq. (14), the Poisson coefficient is independent of the quantity λ_1 only at discrete values of the fractal dimension $d_{\mathrm{f}}^{*} = d - 1$, acquiring here the constant value $\nu = 0$. It is significant that in the case of volume deformation ($d = 3$) this result corresponds to the ideal entropy elasticity of elastomers which is described by potential (1). In any case, the quantity d_{f}^{*} is the upper limit of the values of the dimension for self-similar structures that possess a negative Poisson coefficient.

NOTATION

W, highly elastic potential, J; λ, deformation multiplicity; A and B, material constants, J; ν, Poisson coefficient; d, space dimension; d_f, fractal dimension; ε, relative deformation ($\varepsilon = \lambda - 1$); V, volume, m^3; l and L, linear dimensions, m. Subscripts: $\|$, longitudinal; $\perp$, transverse; f, fractal; 0, undeformed state; i and j, coordinate axes; min, minimum; *, integral values.

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