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THERMOELASTIC PARAMETERS OF LAYERED COMPOSITES

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Abstract. The paper presents calculations of thermoelastic parameters of layered composite structure (substrate + adhesive) using the contact layer method. The corresponding mathematical model (taking into account the presence of a contact layer) has been used to study properties of a layered rod subjected to heating. Temperature dependences of Young's modulus, CLTE and thermal stresses for layered, built-up and polymeric specimens were obtained and analyzed. A comparison of the calculation data on the effective properties of objects under investigation obtained by the classical formulae and by the contact layer method was made. The importance of taking into account the contact layer presence and its parameters in the study of thermoelastic characteristics of layered structures was proved.

Keywords: layered composite, adhesive mechanics, contact layer method, Young's modulus, CLTE

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ТЕРМОУПРУГИЕ ПАРАМЕТРЫ СЛОИСТЫХ КОМПОЗИТОВ

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Аннотация. В статье представлены расчеты термоупругих параметров слоистой композиционной структуры (субстрат + адгезив) по методу контактного слоя. Соответствующая математическая модель (учитывает наличие контактного слоя) использована для исследования свойств слоистого стержня, подвергнутого нагреву. Получены и проанализированы температурные зависимости модуля Юнга, КЛТР и термических напряжений для слоистого, составного и полимерного стержней. Для объектов исследования проведено сравнение результатов расчета их эффективных свойств, полученных по классическим формулам смеси и по методу контактного слоя. Доказана важность учета наличия контактного слоя и его параметров при изучении термоупругих характеристик слоистых структур.

Ключевые слова: слоистый композит, адгезионная механика, метод контактного слоя, модуль Юнга, КЛТР

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Introduction

Over the past few decades, researchers around the world have been developing new composite materials, categorizing materials with a layered structure as a separate class. This structure allows to achieve the required properties of the material, making it relatively easy to manufacture. Layered composites (LCs) are used in many industries for production of structural elements for various purposes [1]. Examples of such elements are rods, plates and shells. Due to their special thermoelastic properties, high strength, corrosion resistance and relatively light weight, composites are promising competitors to traditional materials.

It is crucial to be able to model the physical and mechanical behavior of the material used to obtain reliable results from structural calculations. Much attention is focused on perfecting the physical and mathematical methods for calculating the effective properties of composite materials [2]. As more and more adhesive bonded materials are synthesized, along with novel adhesives, more insights are gained into the mechanism of adhesion and the theories describing it [3]. The mechanical properties of LC structures can vary depending on the geometric and physical parameters of the model, such as sample dimensions, Young's modulus, Poisson's ratio and other parameters of adhesive and substrate materials. Pressure, temperature, aging and external load have a great influence on the parameters of the composite. Thus, to calculate the properties of this type of material, it is extremely important to use the most suitable mathematical models whose accuracy has been confirmed by physical experiments.

Materials and methods

The classical theory introduces the mixture rules, used to estimate the elastic parameters of composites; these formulas were obtained by Voigt (in 1889) [4] and Reiss (in 1929) [5]. The mixture rules for determining the transverse elastic modulus E_{mix} and the effective CLTE α_{mix} are written in the following form:

$$E_{mix} = \frac{E_0 E_1}{V_0 E_1 + V_1 E_0}, \tag{1}$$

$$\alpha_{mix} = \alpha_0 V_0 + \alpha_1 V_1, \tag{2}$$

where the subscripts 0 and 1 correspond to two different materials.

These formulas do not take into account Poisson's ratio and the characteristics of the contact layer resulting from adhesive interaction of materials that are part of the composite and in contact with each other. The mixture rules allow to calculate the thermoelastic parameters only in the first approximation. Numerous studies by Russian [6–10, 12] and foreign [11, 13-19] researchers offer various methods for assessing the stress-strain state and determining the elastic parameters of composites, but the results of calculations based on these methods are not consistent with all experiments and require improvement.

The total contact surface area of the interacting layers increases relative to the volume of the material in calculations of the materials that consist of a relatively large number of layers, which is why it is essential to take into account the adhesive interaction of the layers.

Contact layer model. Turusov proposed a contact layer model with a theoretical justification [20]. This model is based on the hypothesis that there exists a certain layer that is a brush with short elastic bonded rods oriented normally to the contact surface, located between the adhesive and the substrate (Fig. 1). There is no direct contact between the rods in the contact layer and, therefore, there are no normal stresses σ_x and σ_z. Short rods absorb shear stresses σ_{yx}, σ_{zy}, σ_{xz} and, naturally, normal stresses σ_y. If contact problems of mechanics are solved by the known methods from elasticity theory, a singularity of type $x^{-1/2}$ occurs for $x \to 0$, i.e., infinity, occurs at the boundaries between layers and in the corner points near the ends of the contact layer for tangential stresses.

The contact layer model proposed by Turusov allows to satisfy all boundary conditions imposed to solve the Cauchy problem, since it provides a way to avoid infinite stresses at the corner points of the adhesive joint. As such singularities are eliminated, it becomes possible to apply the well-known criteria for the strength of adhesive joints. This model of the contact layer can be assumed to be correct from the standpoint of physical rigor, since the existing theory and indirect experiment suggest that the number of bonds per 1 cm^2 is 10^{12} with adhesive contact, i.e., every tenth atom participates in the interaction of the substrate with the adhesive, which means that these contacts are relatively few and do not touch each other.

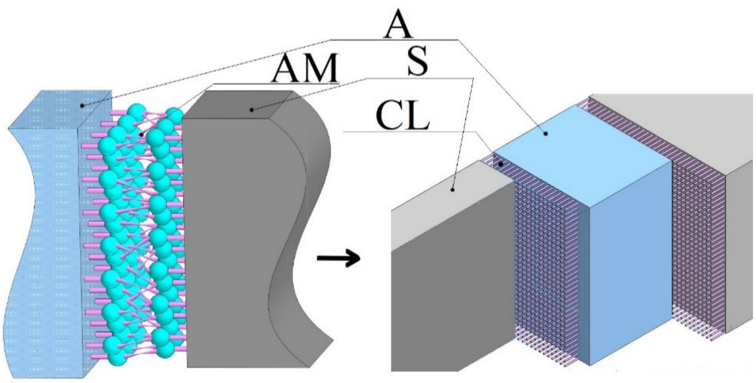

Fig. 1. Schematic representation of the adhesive bond between layers of dissimilar materials brought into contact: A is the adhesive (for example, epoxy), AM are adhesive molecules, S is the substrate (for example, steel), CL is the contact layer

Thus, the contacts together comprise a regular layer of bonded rods, which has certain parameters:

E^* is the elastic modulus, G^* is the shear modulus, h^* is the thickness of the layer.

If the number of bonds per 1 cm^2 is 10^{12}, then every hundredth atom participates in the interaction of the substrate with the adhesive, and in this case the medium parameters are $0.01 \cdot E^*$, $0.01 \cdot G^*$ and h^*.

In accordance with the theory of the contact layer method, Turusov [20] obtained the following relation between the total strain of a layered rod $\varepsilon_{z\,l.b.}$ and its parameters:

$$\varepsilon_{z.l.b.} = \overline{e_z} + \overline{\varepsilon_t} = \frac{\sigma}{E_{eff.l.b.}} + \alpha_{eff.l.b.}\Delta T =$$
$$= q\left[\left(\frac{v_0}{E_0} + \frac{v_1}{E_1}\right) + \frac{2\delta}{\omega^2}\left(\frac{\mu_0 v_0}{E_0 h_0} - \frac{\mu_1 v_1}{E_1 h_1}\right)\left(1 - \frac{th(v)}{v}\right)\right] + \tag{3}$$
$$+\left[\left(\alpha_0 v_0 + \alpha_1 v_1\right) + \frac{2\beta}{\omega^2}\left(\frac{\mu_0 v_0}{E_0 h_0} - \frac{\mu_1 v_1}{E_1 h_1}\right)\left(1 - \frac{th(v)}{v}\right)\right]\Delta T,$$

where the subscripts 0 correspond to the substrate and 1 to the adhesive; E_0, E_1, Pa, are Young's moduli; μ_0, μ_1 are Poisson's coefficients; h_0, h_1, mm, are the thicknesses; V_0, V_1 are the relative volume fractions of the substrate and adhesive content, respectively; e_z is the elastic strain; ε_t is the thermal strain; ΔT, K, is the temperature difference.

The parameters ν, δ, β in the relation (3) characterize the contact layer and are determined as follows:

$$\nu = \frac{\omega l}{2};\ \delta = 2\left(\frac{\mu_1}{E_1} - \frac{\mu_0}{E_0}\right) gr;\ \beta = 2(\alpha_0 - \alpha_1) gr, \tag{4}$$

where l, mm, is the size of the square cross-section side (the length of the layer with the adhesive joint); gr, Pa, is the stiffness of the contact layer;

$$\omega^2 = gr\left[\frac{1-\mu_0}{E_0 h_0} + \frac{2(1-\mu_0)}{E_1 h_1}\right].$$

If we assume that $\Delta T = 0$ in relation (3), then we can derive from it an expression for finding the effective (*eff*) elastic modulus $E_{eff.l.b.}$ (Young's modulus) of a layered rod (*l.b.*), which takes the following form:

$$E_{eff.l.b.} = \left[\left(\frac{V_0}{E_0} + \frac{V_1}{E_1}\right) - \frac{2\cdot\left(\frac{\mu_0}{E_0} - \frac{\mu_1}{E_1}\right)^2}{\frac{(1-\mu_1)}{E_1\cdot V_1} + \frac{(1-\mu_0)}{E_0\cdot V_0}}\cdot\left(1 - \frac{\tanh(\nu)}{\nu}\right)\right]^{-1}. \tag{5}$$

Furthermore, expression (3) can be used to obtain a formula for finding the CLTE $\alpha_{eff.l.b.}$, provided that $q = 0$:

$$\alpha_{eff.l.b} = (\alpha_0\cdot V_0 + \alpha_1\cdot V_1) + \frac{4\cdot(\alpha_0-\alpha_1)\cdot(\mu_0\cdot E_1 - \mu_1\cdot E_0)\cdot V_0\cdot V_1}{(1-\mu_0)\cdot E_1\cdot V_1 + (1-\mu_1)\cdot E_0\cdot V_0}\cdot\left(1 - \frac{\tanh(v)}{v}\right). \tag{6}$$

If we assume that the total strain of the entire layered rod is equal to zero in relation (3), we obtain the following the dependence for the thermal stresses σ in a rod of constant length on various characteristics of the rod:

$$\sigma = -\alpha_{eff.l.b.}\cdot E_{eff.l.b.}\cdot\Delta T. \tag{7}$$

The coefficient of linear thermal expansion (CLTE) is a physical quantity that characterizes the relative variation in the linear dimensions of a body with an increase in temperature by 1 K at constant pressure and without phase transformation. The CLTE is unique for each material and depends on a large number of parameters. This key property of the material turns out to be especially important for studies of composite structures operating in environments with variable temperatures. Crystals typically have the lowest CLTE because their structure is extremely uniform and strong. Solids with the highest CLTE have weak intermolecular bonds; common examples are polymers, known to be characterized by low melting points. Studies into the properties of composite materials containing polymers pay considerable attention to the glass transition temperature, below which the polymer material becomes hard and brittle. This parameter is determined individually for each polymer and depends on the chemical composition and structure of the molecular chain. The transition from a hard and brittle «vitreous» state to a soft and plastic *rubber-like* state occurs when the temperature rises.

Background assumptions for the calculations. As mentioned above, the physical and mechanical characteristics of layered composite materials can be calculated if we know the reliable parameter values of the adhesive, substrate, as well as the characteristics of adhesive interaction of the contacting layers. The study of the thermoelastic behavior of composite rods used the geometric and physical parameters of the specimens from the experiments conducted by Turusov [20].

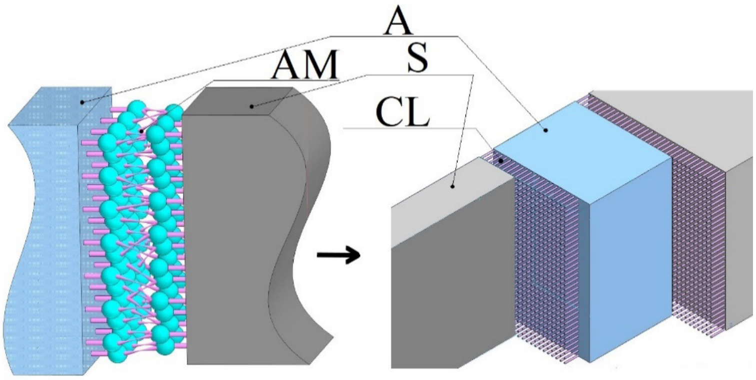

Fig. 1. Schematic representation of the adhesive bond between layers of dissimilar materials brought into contact: A is the adhesive (for example, epoxy), AM are adhesive molecules, S is the substrate (for example, steel), CL is the contact layer

Fig. 2 schematically shows the geometry of the specimens considered. They have the same length; one is made of pure polymer, and the other two consist of two materials: metal and polymer.

Specimen *1* is a rod made of polymer material (in this case, epoxy was taken). Specimen *2* is a rod composed of two steel rod elements (substrates), with a layer of polymer adhesive (epoxy) between them. Specimen *3* is a layered rod made of steel plates (substrates) bonded by epoxy.

Table 1 shows the constant parameters of composites that we have adopted for the calculations. Notably, the layered and composite rods contain the same amount of polymer and steel in their composition (15% and 85% in each of the rods, respectively).

Table 1

Initial data for calculations

Parameter	Notation	Value
Specimen geometry		
Total length, mm	L	100
Size of square section side, mm	L	10.0
Section thickness in specimen *2*, mm steel polymer	 h_{st} h_{pol}	 42.5 15.0
Layer thickness in specimen *3*, mm steel polymer	 h_0 h_1	 1.40 0.25
Physical properties of materials		
Steel: Young's modulus, GPa Poisson's ratio CLTE, K^{-1}	E_0 μ_0 α_0	210 0.3 $1.2\cdot10^{-7}$
Poisson's ratio for polymer	μ_1	0.5
Stiffness of contact layer, GPa/mm	gr	25

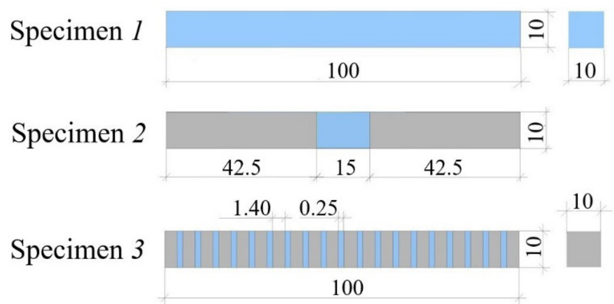

Fig. 2. Geometry of composite rods:
solid polymer (*1*), composite (*2*) and layered (*3*) (dimensions are given in mm)

Results and discussion

Fig. 3,*a* shows graphs of temperature dependences of Young's moduli for polymer, composite and layered rods. Young's modulus of the composite rod (specimen *2*) was calculated by mixture rule (1), and the layered rod (specimen *3*) by the Turusov model (see Eq. (5)). Young's modulus of the polymer E_1 was calculated using an empirical formula obtained by approximating the experimental data measured during a laboratory study of the dependence of Young's modulus of epoxy adhesive E_1 on temperature T [20]. The formula has the following form:

$$E_1 = -18.2 \cdot T + 8200. \tag{8}$$

The following empirical formula was also used for the CLTE α_1 of the polymer:

$$\alpha_1 = (0.46T - 58) \cdot 10^{-6}. \tag{9}$$

The behavior of curve *1* in Fig. 3,*a* shows that the elastic modulus of the polymer decreases almost linearly with an increase in temperature. A more accurate estimate of the difference between the temperature dependences of the elastic modulus can be obtained from analysis of the data in Table 2. The analysis indicates that the elastic modulus of the polymer is 2831 MPa at the initial temperature of the polymer, equal to 295 K, and 1011 MPa at a temperature of 395 K (decreases approximately by 2.8 times with an increase in temperature by 100 K). This decrease in the elastic modulus can be explained by an increase in interatomic and intermolecular distances, as well as a weakening of the interaction forces between microparticles in the bulk of the material.

Table 2

Calculated temperature dependences of elastic modulus and CLTE for the three specimens considered

Temperature, K	Young's modulus, MPa			Thermal expansion coefficient, 10^{-5} K^{-1}		
	1	2	3	*1*	2	3
295	2831	17.534	101.793	7.8	2.19	5.805
305	2.649	16.482	99.065	8.2	2.25	6.134
315	2.467	15.420	96.132	8.7	2.32	6.465
325	2.285	14.349	92.965	9.2	2.39	6.797
335	2.103	13.267	89.535	9.6	2.46	7.130
345	1.921	12.176	85.801	10.1	2.53	7.465
355	1.739	11.074	81.718	10.5	2.60	7.801
365	1.557	9.961	77.227	11.0	2.67	8.138
375	1375	8.839	72.255	11.5	2.74	8.477
385	1193	7.705	66.711	11.9	2.81	8.818
395	1011	6.561	60.475	12.4	2.88	9.161

Note. The specimen numbers correspond to those shown in Fig. 2.

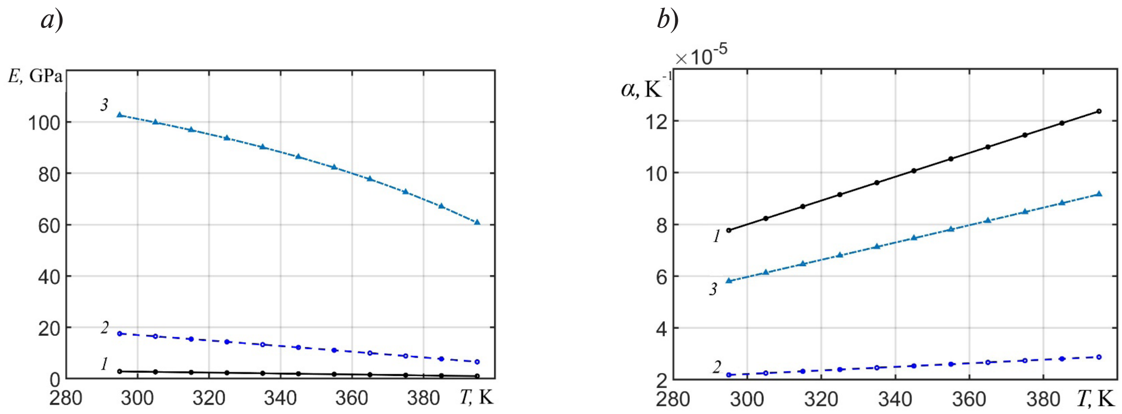

Fig. 3. Calculated effect of temperature on the elastic moduli (*a*) and CLTE (*b*) of polymer (*1*), composite (*2*) and layered (*3*) rods.
Data from Table 1, as well as Eqs. (8) and (9) were used

The results obtained by the mixture rule for the composite rod (see curve *2* in Fig. 3,*a*) indicate that Young's modulus decreases with increasing temperature in similarly to that in the polymer, but curve *2* has a much shallower slope to abscissa than curve *1*. The reason for this difference is that the composite rod (specimen *2*) includes a polymer section between two sections of steel (the relative volume fractions of polymer and steel are 15% and 85%, respectively), which is a consequence of the fact that the initial value of Young's modulus of such a composite rod is greater than that of the polymer. The elastic modulus decreases by 2.7 times with an increase in temperature by 100 K.

Curve *3* in Fig. 3,*a*, obtained by Turusov's formula, is markedly different from the first two. It is important to note for this case that the value of Young's modulus at the initial temperature obtained by Eq. (5) is much higher than that obtained by the mixture rule. Such a synergistic effect in the layered structure is due to the influence of the contact layer that appears between the adhesive and the substrate. Expression (4) allows taking into account the presence of the contact layer and its parameters, which is what produces the observed discrepancy. Since the calculation was carried out for a soft polymer (adhesive), whose Young's modulus is significantly higher than Young's modulus of steel (substrate), we introduce here the bulk modulus *K* for uniform stretching (instead of the standard elastic modulus of the polymer), depending on both the elastic modulus and Poisson's ratio:

$$K = \frac{E}{2(1-2\mu)}. \tag{10}$$

Poisson's ratio is close to 0.5 in the above calculation, which means that the modulus *K* is superior to Young's modulus and the resistance of the layered structure to stretching and compression.

Considering the properties of the materials, we found that the polymer has a significant fraction of free volume, for example, cavities of the order of molecular (monomeric) sizes or voids of smaller magnitude associated with irregular packing of molecules. The process of thermal expansion of the polymer is mainly an increase in the free volume with an increase in temperature, while similar processes in crystalline solids, characterized by much lower values of CLTE, are associated with anharmonic dependence of potential energy on interatomic or intermolecular distances [22]. Rupture stresses can occur in composite structures that are layered materials, for example, due to the difference in the coefficients of linear thermal expansion of the components that make up the layered composite.

Fig. 3,*b* shows the temperature dependences of CLTE for polymer, composite and layered rods. Evidently, all three curves exhibit growth with increasing temperature. The CLTE of the polymer increases because the molecules become more mobile as the temperature increases, weakening intermolecular bonds during motion.

According to the calculated data for the layered rod, the presence of contact layers significantly affects the results; this is a consequence of the predominant influence of the bulk modulus K for uniform stretching (instead of standard Young's modulus of the polymer, as already mentioned above); as a result, the CLTE of the layered rod takes a greater value than that of the composite, calculated by the mixture rule.

In practice, to avoid delamination in materials and destruction of their structure, it is of utmost importance to correctly calculate the thermal stresses in composite materials. Our calculations of thermal stresses in three specimens show that the behavior of the layered composite with an increase in temperature is radically different from the corresponding reactions of composite (three-section) and polymer rods.

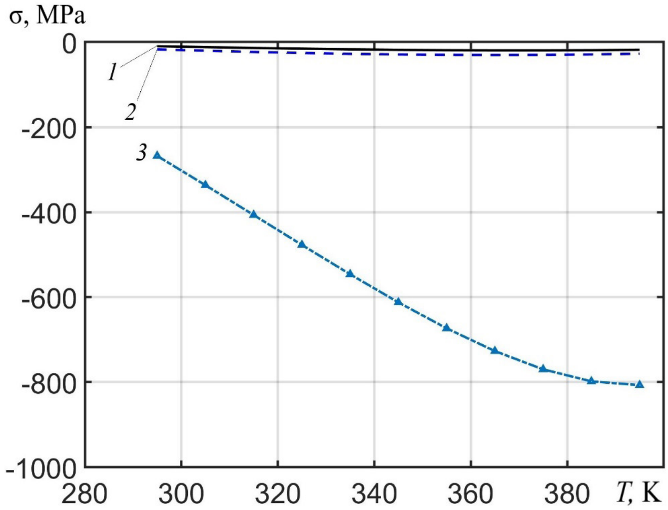

Fig. 4. Temperature dependences of thermal stresses in polymer (*1*), composite (*2*) and layered (*3*) rods (curves *1* and *2* almost coincided)
The data from Table 1, as well as Eqs. (8), (9) were used

Table 3

Calculated temperature dependences of heating-induced stresses in three specimens

Temperature, K	Stress σ, MPa		
	1	*2*	*3*
295	–9.899	–17.244	–267.866
305	–11.991	–20.437	–336.650
315	–13.935	–23.289	–406.812
325	–15.681	–25.747	–477.141
335	–17.178	–27.758	–546.201
345	–18.377	–29.270	–612.286
355	–19.227	–30.225	–673.335
365	–19.678	–30.570	–726.839
375	–19.680	–30.245	–769.702
385	–19.182	–29.194	–798.035
395	–18.134	–27.356	–806.866
405	–16.486	–24.672	–789.686
415	–14.188	–21.079	–737.708

Note. The specimen numbers correspond to those shown in Fig. 2.

Based on the values obtained (Table 3) and the constructed graphs (Fig. 4), we can conclude that the thermal stresses in the polymer and composite rods are much less than those in the layered rod. The reason for this is that the values of Young's modulus and CLTE obtained by the mixture rule are less than those obtained by the formula for the layered rod. Data analysis suggests that the stresses increased in absolute magnitude at temperatures from 300 to 400 K, but began to decrease in all three cases at 380 K. A more intense growth in the CLTE (relative to the initial value) with an increase in temperature, compared with the decrease rate of the elastic modulus, indicates a decrease in thermal stresses at a temperature of 380 K.

We should emphasize that the CLTE value of the polymer is much higher than the corresponding value for steel (see Table 1). Polymer and steel layers in the layered and composite rods were rigidly connected to each other and subjected to heating; each material tends to expand in accordance with its CLTE value, but since it is higher in polymer and lower in steel, thermal stresses arise (the polymer tends to expand, and steel prevents this). The steel is subjected to tensile stress, since the polymer forces it to expand beyond the limit that is defined by its CLTE value. Stresses arise due to the difference between the adhesive (epoxy) and the substrate (steel) and can lead to delamination. We can observe from the above dependences that the temperature variation in the entire range under consideration changes the value of the CLTE for the polymer by more than six times, and the elastic modulus drops to zero as the temperature approaches the melting point. Such dependencies are also true for steel, although to a lesser extent.

Conclusion

The paper reports on the calculations of thermoelastic properties of layered composites, analyzing the data obtained. The physical mechanism of the transformations in the material exposed to temperature variation was clarified in the analysis. Calculations of Young's modulus and the coefficient of linear thermal expansion (CLTE) were carried out based on the theory of the contact layer method for a layered rod and in accordance with the classical mixture rule for a composite one. The theory of the contact layer method allows to take into account the geometric and physico-mechanical parameters of the substrate, adhesive and contact layer (the layer formed by their interaction), providing better accuracy for the simulations and calculations.

We established that calculations of the characteristics of a layered rod should focus closely on the adhesive interaction of interfacial layers. An evident conclusion from the calculations and analysis of the results is that a large number of mechanical characteristics should be taken into account in studies into the thermoelastic parameters of a rod with a layered structure. In particular, Poisson's ratio, the thickness of the component layers and the stiffness of the contact layer have a significant effect.

The coefficients of linear thermal expansion were calculated using the contact layer method and the mixture formula.

Importantly, the analytical formula (5) for determining effective Young's modulus by the contact layer model agrees fairly well with the results of the physical experiment and can be considered fairly accurate. The effective modulus taking into account the contact layer as a parameter allows to conduct numerical experiments by varying the values of the mechanical properties of the materials that make up the layered composite.

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