



MODELING OF STRUCTURIZATION IN THE ADHESIVE LAYER IN THE CREATION OF MULTILAYER OBJECTS

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In this paper, we propose a thermophysical model of structurization in the adhesive layer in the process of creating products by the lamination. Modeling is reduced to the solution of a three-layer conjugate thermal conductivity problem. The thermal contact between the layers is assumed to be ideal. It is assumed that structurization activated in the intermediate (adhesive) layer can lead to a change in the thermal conductivity coefficient of the adhesive. The lamination process is controlled by a hot roll with a given radius and a fixed temperature. The roll moves along the outer surface at a constant velocity; the force of the roll pressing on the surface determines the size of the contact area and the maximum effective stresses, which are known from the solution of the contact problem and are the parameters in the proposed model. The structurization is represented as a reversible reaction, the forward and reverse rates of which depend on the temperature and the acting stresses. The problem is implemented numerically. The thermal problem is solved using the difference scheme of splitting in coordinates and a double-sweep method; the kinetic problem is solved using the semi-implicit Euler method. The fields of temperature and structurization level are calculated at different points of time by varying the values of model parameters. To find stationary or quasi-stationary modes of the structurization process, the behavior of the average integral values of the structurization level and temperature is investigated. It was found that the model parameters have an ambiguous effect on the nature of the temperature distribution and the structurization level; the structurization process does not proceed to its end. The structurization level in the adhesive layer depends on the process parameters (roll temperature, applied pressure and velocity of its movement), as well as on the geometric and physical parameters of the layers.

Key words: laminated object manufacturing, three-layer system, structurization, adhesive layer, thermophysical model

1. Introduction

Laminated Object Manufacturing (Laminated Object Manufacturing or Sheet Lamination - LOM) refers to the widespread technologies for producing three-dimensional objects. If initially LOM was used in the production of paper with a protective layer, then nowadays this technology is used in such fields as construction, microelectronics, production of ceramic/glass-ceramic objects, various kinds of composites, organic products, foil bonding and others. The variety of LOM implementation is due to the ease of its use and high yield of the final product [1, 2].

In the process of creating a three-dimensional object by LOM method, 4 stages are conventionally distinguished. The first stage consists in the development with the help of CAD system of the future product model - CAD model, which is loaded into the industrial computer of the installation. At the second stage, sheet material with an adhesive layer pre-applied between the sheets is fed to the working platform of the machine. The third stage involves cutting out the contours of the manufactured product with a laser. At the fourth stage, the resulting sheets are bonded in layers by rolling a heated roll, and removing excess consumables.

Despite the significant advantages of the technology, there are problems that need to be solved. The LOM method works well when joining sheets of paper, plastic, organic. High production accuracy and easy bonding of the same type of materials is achieved by varying the thickness of their layers in the range from 0.05 to 0.1 mm. However, the replacement of one of them, for example, metal, is reflected as an increase in the thickness of the sheet from 0.2 to 0.5 mm, which leads to a complication of the gluing operation. Then, for example, as in [3], a two-step process is used to bond metal foils. First, the sheets of metal foil are shaped using LOM, and then the layers are joined by diffusion welding. Another problem of foil joining is the jagged boundaries of metal functional products with complex shapes and a large volume of cut material. This problem is discussed in detail in the article [4], in which a theoretical study on improving the accuracy of LOM technology for the production of complex-shaped products was conducted. The authors obtained results that make it possible to influence the production error by changing the cut surface of excess material by means of a laser. Thus, for a quarter-cylinder-shaped product with a radius of 100 mm and a height of 50 mm with a layer thickness of 1

mm it was possible to reduce waste by 3.7 times. In [5], a two-step technological process was carried out to create a functionally graded Ti-C-Ni material. At the first stage LOM technology was used to join metal sheets, at the second stage - combustion synthesis. This allowed the authors to achieve a continuous distribution of the Ti+C+Ni bond in the volume of the material, which resulted in a homogeneous distribution of properties.

Physical and chemical processes can be observed in the bonding compositions when obtaining laminated products by the LOM method. Thus, traditionally used for this purpose organic materials and adhesives of different compositions undergo phase transitions and structural changes that contribute to the formation of the connection. Since the synthesis process in the adhesion layer is accompanied by phenomena of physical nature of different directions and with different spatial and temporal scales, critical conditions that are necessary for its initiation are possible. Direct observation of structural changes is not possible. In order to study and explain the regularities that lead to the formation of multilayered objects, it is necessary to resort to mathematical modeling. Prototypical models of the processes that accompany LOM can be models of macrokinetics, which describe the modes of exothermic reactions in layered systems. For example, in [6], combustion in a system consisting of alternating layers of a combustible and an inert layer of given thicknesses is considered. The reaction in the combustible proceeds without the participation of the gas phase, and the thermophysical properties of the layers are unchanged. A conditional correlation of combustion in a model system and in a real heterogeneous material is proposed. In [7], the role of competing heat transfer mechanisms of conduction and radiation in establishing different combustion regimes is discussed.

In [8, 9], the peculiarities of exothermic reaction propagation between layers of inert material are investigated. The combustion temperature in the model is controlled by diluting the exothermic composition with inert particles, the heating of which consumes part of the heat. Publications [10-12] are related to the study of compound formation of heterogeneous materials; the effect of differences in the thermal properties of all materials on the combustion rate and the maximum temperature at the front is analyzed.

Work [13] is devoted to combustion in two-layer systems consisting of low-melting metals or alloys and high-energy mixtures with the formation of porous products. As a result, a composite material layer is formed between the layers, the thickness of which is determined by the depth of melt penetration into the porous combustion product. Burning of the layered package is investigated in [14]. Some layers are high-calorie compositions, while the other are inert or low-calorie compositions. For both systems, the authors identified stationary and non-stationary combustion modes and showed the possibility of transition between modes. In [15], combustion of a layered composition of two porous reaction layers separated by a layer of a fusible inert material was analyzed. The combustion front moves in the direction perpendicular to the position of the layers, and the possibility of a concomitant process of impregnation of the porous layer with the melt was shown.

In [16], a model of exothermic chemical reaction propagation in the layer between two inert materials with different properties is proposed, provided that a heat source of finite dimensions moves along the outer surface. In fact, the synthesis of the bonding compound directly in the lamination process is modeled. Calculation results showing the existence of different process regimes are presented.

It should be noted that exothermically reacting compositions of bonding layers are not yet widespread in LOM technology. In most cases, compositions are used that undergo some structural changes (structural phase transitions, polymerization, recrystallization, compaction and others) during the heating process combined with mechanical loading, which are not accompanied by significant thermal effects. When describing the process at the macro level and for all such processes, it is convenient to apply a single parameter. The authors of this work did not find a generally accepted term in the literature. Different researchers refer to the same phenomena as "structural transformations", "structurization", "structurization", "structure formation", "structure formation", and the corresponding value - "degree of structurization", "degree of structurization", "degree of transformation or depth of transformation".

In this article, we will use the terms "structurization" and "degree of structurization" to macroscopically describe the process in order to distinguish it from the degree of transformation in reactions accompanied by significant thermal effects. A thermokinetic model of structurization in the adhesive layer leading to a change in the properties of this layer is formulated for the first time. Depending on the temperature of the adhesion regulating roll, its speed and pressing pressure, different conditions for structure formation are realized. The contact spot area and maximum stresses follow from the solution of the known contact problem. Thus, the aim of the work is to study the role of controlling factors, such as the pressure of the roll, its temperature and speed of its movement, in establishing the stationary or quasi-stationary mode of laminating.

2. Formulation of the problem

Consider a three-layer system consisting of two inert layers and an adhesion layer between them, as shown in Figure 1. To connect layers *A* and *B*, a roll of radius R_0 , heated to temperature T_s , moves along the surface of the upper layer at a constant velocity v . The width of the roll coincides with the width of the sample.

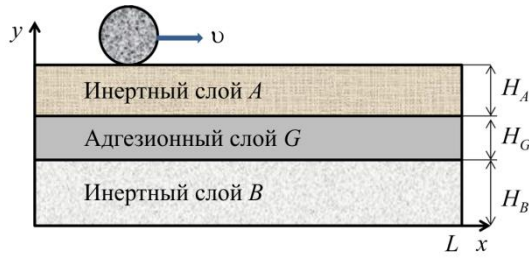


Fig. 1. Problem Scheme.

The maximum stress in the contact zone arising when pressure is applied to the roll moving on the surface of the inert layer *A*, is [17]:

$$\sigma_{\max} = \mu \sqrt{\frac{p}{2R_0(\theta_1 + \theta_2)}}, \quad (1)$$

and the half-width of the contact area is found by the formula:

$$s = \mu \sqrt{2pR_0(\theta_1 + \theta_2)}. \quad (2)$$

Here: $\mu = 0,798$ — dimensional coefficient, following from the approximate solution of the contact problem; p — roll pressure; $\theta_k = (1 - \nu_k)/E_k$ — complexes including Poisson's coefficients ν_k and elastic moduli E_k of contacting bodies ($k=1,2$). It is this stress that is transferred to the whole sample and influences the processes in the adhesion layer.

As a result, the determination of the temperature field is reduced to the solution of a two-dimensional coupled thermal conductivity problem, including the thermal conductivity equations for the inert layers:

$$c_k \rho_k \frac{\partial T_k}{\partial t} = \lambda_k \left(\frac{\partial^2 T_k}{\partial x^2} + \frac{\partial^2 T_k}{\partial y^2} \right), \quad (3)$$

where index k describes inert layers *A* and *B*, c_k — the heat capacity, ρ_k — the density, T_k — is the temperature, λ_k — is the thermal conductivity coefficient, x and y — spatial coordinates, t — is the time.

Depending on the type of materials to be joined and the expected properties of the laminated composite, different adhesion materials (powders, films, organic and inorganic materials) are taken, in which chemical reactions and various structural changes can be observed: phase transitions, polymerization, recrystallization, compaction and others. Let us assume that there are no chemical reactions leading to a change in the composition in the adhesion layer, but restructuring of its material structure is possible. To separate this process from the one caused by the chemical reaction itself, we will call it structurization. This makes the model in this work different from the one used in [16].

According to [18-21], the process of structural change can be represented as a reversible reaction that is not accompanied by any significant thermal effects. Then the degree of structural transformations a will follow from the kinetic equation:

$$\frac{da}{dt} = w_2 - w_1, \quad (4)$$

where $w_2 = k_2(1-a)$ and $w_1 = k_1a$, are the forward and reverse reaction rates, respectively. Under nonisothermal conditions, k_2 , k_1 are functions of temperature. All of the above processes are thermally activated, and their rates depend on the temperature according to the Arrhenius law. In the case of reactions involving solids, the law for the reaction rate may be more complicated [22, 23]. It is possible to activate a reaction not only by changing the temperature (internal energy), but also by doing work [24]. This corresponds to another exponential law. For example, in solid-state chemistry, the Eyring-Kozman formula is widely used

$$w \sim k_0 \exp\left(\frac{E - \sigma V^\ddagger}{RT}\right),$$

where V^\ddagger — activation volume, σ — acting stress, k_0 — pre-exponential factor, E — activation energy, R — universal gas constant, T — temperature. In writing this equation, it is assumed that the acting stresses simplify the transition through the energy barrier, but do not affect its height [25].

Note that the detailed scheme of transformations (reaction stages), as a rule, is not clear or is complex and ambiguous. The rate of transformations in the solid phase is determined not only by chemical reactions, but also by numerous physical processes that are difficult to study directly; these processes take place at the microlevel (at the level of individual particles, grains, interfaces). The easiest way out in this situation is to use such a macroscopic concept as the conversion level (conversion degree) [22-25]. This is exactly what is done in macrokinetics [26]. A kinetic function appears in the laws for the rate of reactions, the form of which depends on the processes at the microlevel. In addition, almost any reaction can potentially go forward as well as backward. This is true for all of these processes. According to experimental data [3-5, 27, 28], the structure is not completely formed during a single roll pass, which is partly due to the reversibility of this process and partly due to the action of competing factors. The combination of temperature and mechanical loading makes it possible to control the process. As a result, for the rates of the forward and reverse processes we write:

$$w_1 = k_1a = k_{10}a \exp\left(\alpha_1 \sigma_{\max} - \frac{E_1}{RT_G}\right), \quad w_2 = k_2(1-a) = k_{20}(1-a) \exp\left(\alpha_2 \sigma_{\max} - \frac{E_2}{RT_G}\right).$$

Here: k_{10} , k_{20} — pre-exponential factors; α_1 , α_2 — the sensitivity coefficients of the reaction rates to the acting stress; E_2 , E_1 — activation energy of the forward and reverse structurization stages; R — universal gas constant; T_G — температура адгезионного слоя; a — level of structural transformations (degree of structurization).

Note that even in relatively studied processes, specific kinetic parameters require special experimental determination, which is not the purpose of this work. However, it is known that structurization can lead to changes in the properties [18-21]. Thus, according to [29], the thermal conductivity coefficient of amorphous and partially crystallized polymers may be different. The thermal conductivity coefficients of cardboard of different structures differ by almost an order of magnitude [30]. The same order of divergence of thermal conductivity coefficients of dry and tamped soil. In the problem considered in this paper, it is the thermal conductivity that is of the greatest interest, since the level of inhomogeneity of temperature distribution and, as a result, the nature of structurization depends on the ratio of thermal conductivities of the layers. Let us assume that structurization will cause a change in the thermal conductivity coefficient of the adhesion layer: $\lambda_G = \lambda_2(1-a) + \lambda_1a$ where λ_2 and λ_1 are thermal conductivity coefficients of unstructured (index 2) and fully structured (1) adhesion layers. It is possible to use only linear dependence on the composition (or mixture rule), since the specific structure at the microlevel is not known within the framework of this model. So, there is either reagent $(1-a)$, or product (a) , or mixture of them. Then the thermal conductivity equation for the adhesion layer will take the form:

$$c_G \rho_G \frac{\partial T_G}{\partial t} = \lambda_2 \frac{\partial}{\partial x} \left(\left(1 + a \left(\frac{\lambda_1}{\lambda_2} - 1 \right) \right) \frac{\partial T_G}{\partial x} \right) + \lambda_2 \frac{\partial}{\partial y} \left(\left(1 + a \left(\frac{\lambda_1}{\lambda_2} - 1 \right) \right) \frac{\partial T_G}{\partial y} \right). \quad (5)$$

Let us take the adiabatic conditions at the ends and in the plane $y = 0$:

$$x=0, \quad x=L: \quad \lambda_k \frac{\partial T_k}{\partial x} = 0 \quad (k = B, G, A), \quad (6)$$

$$y=0: \quad \lambda_B \frac{\partial T_B}{\partial y} = 0. \quad (7)$$

At the interfaces between the layers, let us assume that the conditions of ideal thermal contact are true:

$$y = H_B: \quad \lambda_B \frac{\partial T_B}{\partial y} = (\lambda_2(1-a) + \lambda_1 a) \frac{\partial T_G}{\partial y}, \quad T_B = T_G, \quad (8)$$

$$y = H_B + H_G: \quad (\lambda_2(1-a) + \lambda_1 a) \frac{\partial T_G}{\partial y} = \lambda_A \frac{\partial T_A}{\partial y}, \quad T_G = T_A. \quad (9)$$

On the surface of layer A, due to the movement of the hot roll, there is a movable heat source of limited size $2s$. For $y = H_B + H_G + H_A$ we have the following conditions:

– in the contact zone of the roll with the layer A, i.e. at $\nu t + x_0 - s \leq x \leq \nu t + x_0 + s$

$$T_A = T_S; \quad (10)$$

– out of the contact zone, i.e. at $x \leq \nu t + x_0 - s$ and $x \geq \nu t + x_0 + s$

$$\frac{\partial T_A}{\partial y} = 0. \quad (11)$$

At the initial moment the sample has a temperature T_0 , the layer G is unstructured ($a = 0$).

The model contains physical parameters that can be found in the literature, obtained from experiments or calculated on the basis of chemical thermodynamics, but the accuracy of their determination will not be high. In such cases in macrokinetics (i.e., in the study of chemical processes, taking into account the accompanying physical stages), the analysis of models in dimensionless variables is used, which allows to establish the most essential parameters for the process, to identify the criteria characterizing the process. Therefore, let us represent the system (1)-(11) in a dimensionless form.

The analysis of the problem in dimensionless variables is also advantageous because the model presented in the article covers general properties of different technologies, in which material properties, product sizes, and roll rates and temperatures vary considerably. The sizes of objects that are created in LOM technologies vary widely. The possibility of quickly obtaining parts of large size (up to several meters) is one of the advantages of this technology. The roll size and pressing pressure also vary quite widely and depend both on the parameters of the production equipment and on the materials used. For example, in [31], where a simplified thermophysical model was presented, the data for the roll were as follows: $s = 9$ mm; $\nu = 25,4$ mm/s; $T_S = 91$ °C, and thermal-physical properties of ceramics. In [32] technological parameters were varied, loads were set: $p = 12, 25, 40, 51$ kg; roll temperatures: $T_S = 120, 170, 220, 270, 320$ °C; velocities: $\nu = 5, 10, 20, 40$ mm/s. It was assumed that the steel sheets were connected by acrylic glue. Thermocouples were used to measure the temperature in different layers, which made it possible to construct a typical temperature-time curve for problems with moving heat sources. In [33], based on a semi-analytical model, the contact spot size, temperature and stresses in the vicinity of the roll were calculated at its parameters: $T_S = 200$ °C; $\nu = 1,27$ cm/s. The article also states a thermal conductivity of 0.501 W/(mK). In general, the procedure for making metal sheet laminates is similar to that for creating laminated paper. The technology uses organic materials, polymers, ceramics, compressed powder materials and others. Laminates are obtained not only by combining heterogeneous materials, but also by gradually changing the composition and properties of the layers.

Therefore, to find the most general results and parametric study of the model let us go to dimensionless variables:

$$\theta_k = \frac{T_k - T_S}{RT_S^2} E_2, \quad \xi = \frac{x}{H_G}, \quad \eta = \frac{y}{H_G}, \quad \tau = \frac{t}{t_a}, \quad t_a = (k_{20})^{-1} \exp\left(\frac{E_2}{RT_S}\right),$$

where t_a — characteristic time of a direct structurization reaction.

After unmeasuring, the model takes the form:

$$K_{ck} \frac{\partial \theta_k}{\partial \tau} = \frac{K_{\lambda k}}{\delta} \left(\frac{\partial^2 \theta_k}{\partial \xi^2} + \frac{\partial^2 \theta_k}{\partial \eta^2} \right) \quad (k = A, B), \quad (12)$$

$$\frac{\partial \theta_G}{\partial \tau} = \frac{1}{\delta} \left[\frac{\partial}{\partial \xi} \left(((1-a) + \kappa a) \frac{\partial \theta_G}{\partial \xi} \right) + \frac{\partial}{\partial \eta} \left(((1-a) + \kappa a) \frac{\partial \theta_G}{\partial \eta} \right) \right], \quad (13)$$

$$\frac{da}{d\tau} = (1-a)\phi_2(\theta, \sigma) - \gamma a \phi_1(\theta, \sigma). \quad (14)$$

Here

$$\gamma = \frac{k_{10}}{k_{20}}, \quad \phi_1(\theta, \sigma) = \exp\left(\bar{\alpha}_1 \sigma + \frac{\varepsilon + \theta_G}{1 + \beta \theta_G}\right), \quad \phi_2(\theta, \sigma) = \exp\left(\bar{\alpha}_2 \sigma + \frac{\theta_G}{1 + \beta \theta_G}\right).$$

Let us write the boundary conditions:

$$\xi = 0, \quad \xi = \bar{L}: \quad \frac{\partial \theta_k}{\partial \xi} = 0 \quad (k = A, G, B); \quad (15)$$

$$\eta = 0: \quad \frac{\partial \theta_B}{\partial \eta} = 0; \quad (16)$$

$$\eta = \Delta_B: \quad K_{\lambda B} \frac{\partial \theta_B}{\partial \eta} = ((1-a) + \kappa a) \frac{\partial \theta_G}{\partial \eta}, \quad \theta_B = \theta_G; \quad (17)$$

$$\eta = 1 + \Delta_B: \quad ((1-a) + \kappa a) \frac{\partial \theta_G}{\partial \eta} = K_{\lambda A} \frac{\partial \theta_A}{\partial \eta}, \quad \theta_G = \theta_A; \quad (18)$$

$$\eta = 1 + \Delta_B + \Delta_A: \quad \theta_A = 0, \quad \text{if} \quad \omega\tau + \xi_0 - \xi_A \leq \xi \leq \omega\tau + \xi_0 + \xi_A; \quad (19)$$

$$\frac{\partial \theta_A}{\partial \eta} = 0, \quad \text{if} \quad \xi \leq \omega\tau + \xi_0 - \xi_A \quad \text{и} \quad \xi \geq \omega\tau + \xi_0 + \xi_A; \quad (20)$$

and initial conditions ($\tau = 0$):

$$a = 0; \quad \theta_k = -\theta_0, \quad \text{where} \quad k = B, G, A. \quad (21)$$

The problem (12)-(21) includes the following dimensionless quantities:

- geometric parameters (length of the sample, heights of inert layers A and B):

$$\bar{L} = L/H_G, \quad \Delta_A = H_A/H_G, \quad \Delta_B = H_B/H_G;$$

the height of layer G in dimensionless variables becomes one ($\Delta_G = 1$);

- parameters related to the thermal-physical properties of layer materials and structural transformations:

$$K_{ck} = \frac{c_k \rho_k}{c_G \rho_G}, \quad K_{\lambda k} = \frac{\lambda_k}{\lambda_G} \quad (k = B, A),$$

$$\beta = \frac{RT_S}{E_2}, \quad \theta_0 = \frac{T_S - T_0}{RT_S^2} E_2, \quad \delta = \frac{H_G^2 c_G \rho_G}{\lambda_2 t_a},$$

$$\kappa = \frac{\lambda_1}{\lambda_2}, \quad \varepsilon = \frac{E_2 - E_1}{RT_s}, \quad \gamma = \frac{k_{10}}{k_{20}}, \quad \bar{\alpha}_1 = \alpha_1 p_*, \quad \bar{\alpha}_2 = \alpha_2 p_*;$$

– process plant characteristics:

$$\Pi = \frac{p}{p_*}, \quad \xi_A = \sqrt{\Pi}, \quad \sigma = \alpha \sqrt{\Pi}, \quad \xi_0 = \frac{x_0}{H_G}, \quad \omega = \frac{v t_a}{H_G},$$

herewith it is accepted

$$: p_* = \frac{H_G^2}{2\mu^2 R_0 (\theta_1 + \theta_2)}.$$

During the study, the values of dimensionless quantities varied within a fairly wide range. The illustrations are presented for a limited set of their values.

3. Numerical investigation method

For the numerical realization of the problem (12)-(21), we used an implicit difference scheme, the coordinate splitting method, and a linear double-sweep method [34]. The convergence of the numerical solution was checked on the solution of the problem for a moving roll at the parameters: $\beta = 0,1$; $\gamma = 0,2$; $\theta_0 = 5$; $\delta = 50$; $\bar{L} = 24$; $\omega = 0,01$; $\Pi = 1$; $\varepsilon = 0,87$; $\kappa = 0,1$; $\bar{\alpha}_1 = \bar{\alpha}_2 = 0$; $K_{CA} = K_{CB} = K_{\lambda A} = K_{\lambda B} = 1$. As can be seen from Table 1, when changing the steps in space and time, the results of temperature calculation in the observation points diverge by no more than 2%, which confirms the good convergence of the chosen algorithm.

Table 1. Checking the convergence of the numerical solution at $\omega = 0,01$, $\Pi = 1$, $\tau = 1200$

Time step ($\Delta\tau$)	Spatial step ($\Delta\xi = \Delta\eta$)	Temperature value of the sample at the observation points			
		$\theta(0, H/2)$	$\theta(L/4, H/2)$	$\theta(L/3, H/2)$	$\theta(L/2, H/2)$
1,00	0,02	-2,8144	-2,7724	-2,7381	-2,6287
	0,01	-2,8087	-2,7668	-2,7326	-2,6232
	0,005	-2,8073	-2,7655	-2,7313	-2,6221
	0,0025	-2,8065	-2,7648	-2,7306	-2,6215
0,50	0,02	-2,7980	-2,7558	-2,7213	-2,6114
	0,01	-2,7931	-2,7511	-2,7166	-2,6067
	0,005	-2,7900	-2,7480	-2,7136	-2,6037
	0,0025	-2,7891	-2,7472	-2,7128	-2,6030
0,25	0,02	-2,7844	-2,7420	-2,7075	-2,5971
	0,01	-2,7815	-2,7393	-2,7047	-2,5944
	0,005	-2,7787	-2,7366	-2,7020	-2,5917
	0,0025	-2,7770	-2,7349	-2,7003	-2,5900

4. The results of the work

The character of the temperature distribution and the degree of structural transformations in the sample depend on the speed of the roll and the pressure of pressing, as well as on the ratio of the thermo-physical properties of the materials. The temperatures and velocities required to obtain good adhesion joints can differ considerably, and the properties of the layers can differ by a whole order of magnitude. All calculations, the results of which are presented further in Figures 2-5, were performed at $\omega = 0,01$, $\Pi = 1$, $\gamma = 0,5$, $\delta = 5$, $\varepsilon = 0,87$, $\bar{\alpha}_1 = \bar{\alpha}_2 = 0$. The boundaries of the adhesion layer are plotted with dashed lines. In the case of a moving roll (the center of the contact spot is at a point with a variable coordinate), the temperature distribution (see Fig. 2) is heterogeneous even if the thermal properties of layers A and B are the same. At high roll rates, the sample may not have time to warm up to the temperature required for the structurization to begin.

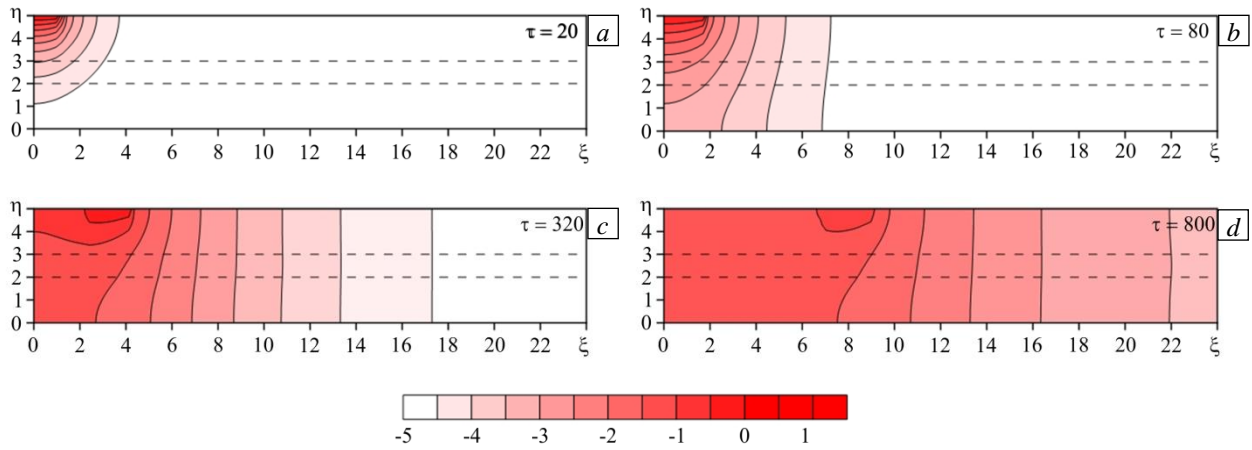


Fig. 2. Two-dimensional temperature distributions in the sample at different moments of time τ at $K_{CA} = K_{CB} = K_{\lambda A} = K_{\lambda B} = 1$; the center of the contact patch is at the point with the coordinate ξ : 0,2 (a); 0,8 (b); 3,2 (c); 8,0 (d).

As the roll advances, the sample gradually warms up. The heterogeneity of heating is clearly visible at the initial stage of the roll movement. For example, at the boundaries of the adhesion layer, the difference in temperature values is recorded up to the time $\tau = 1280$ — the moment when the roll arrives at the point $\xi = 12,8$ (see Fig. 3a). A similar situation is observed with the degree of structural transformations up to $\tau = 320$ (Fig. 3b); at this point, the roll is at $\xi = 3,2$.

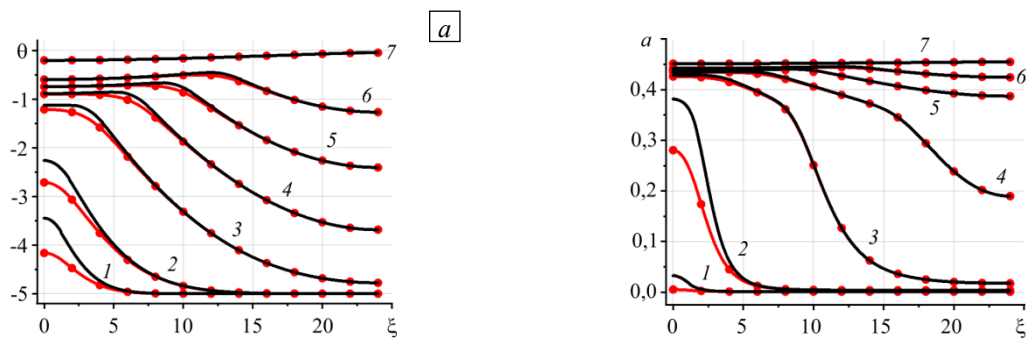


Fig. 3. Temperature distribution (a) and degree of structural transformations (b) on the upper (solid lines) and lower (dotted lines) boundaries of the adhesion layer at different moments of time τ : 80 (curve 1), 80 (2), 320 (3), 640 (4), 960 (5), 1280 (6), 2400 (7).

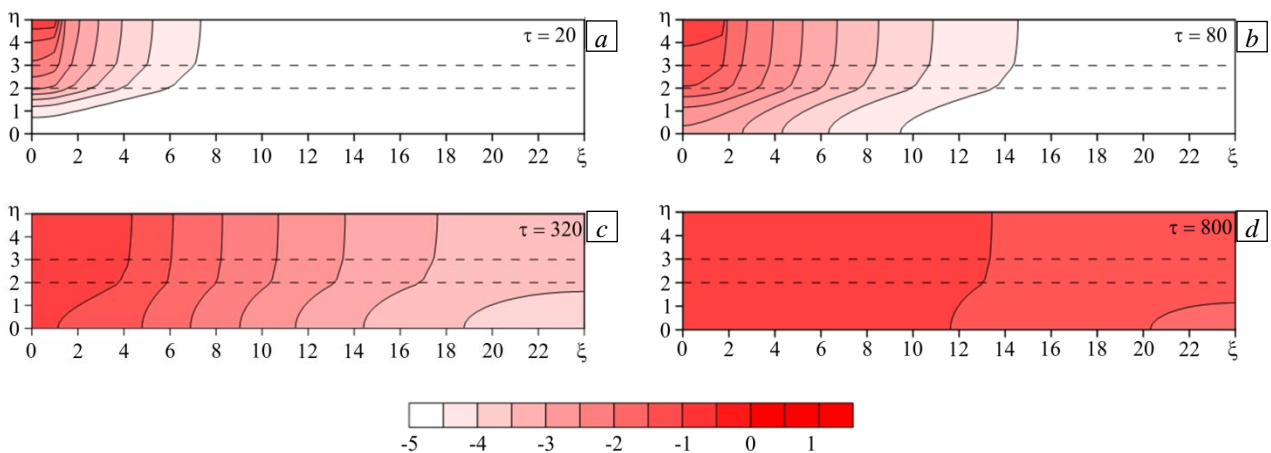


Fig. 4. Two-dimensional temperature distributions in the sample at different moments of time τ for $K_{CA} = K_{CB} = 1, K_{\lambda A} = 5, K_{\lambda B} = 0,2$; the center of the contact patch is at the point with the coordinate ξ : 0,2 (a); 0,8 (b); 3,2 (c); 8,0 (d) заменить кириллицу

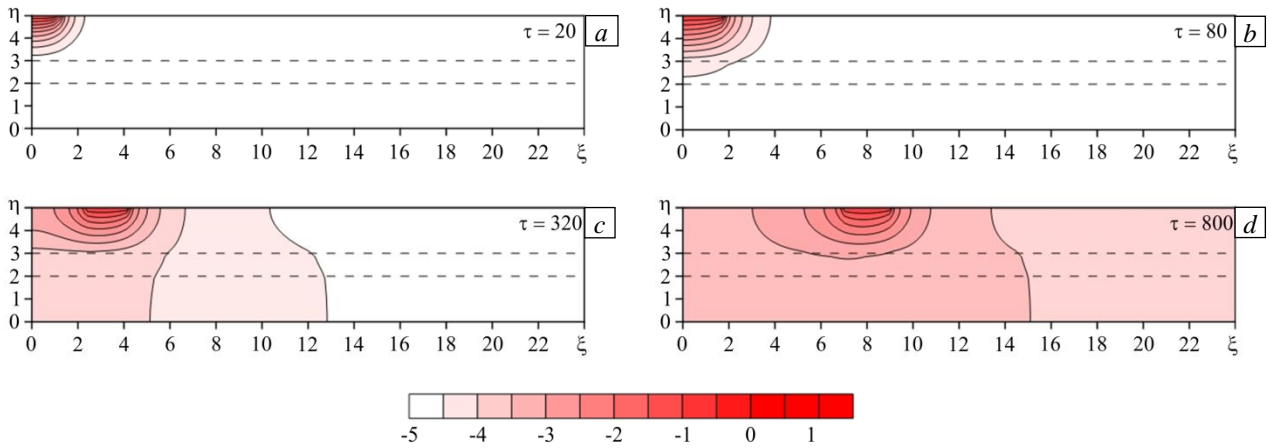


Fig. 5. Two-dimensional temperature distributions in the sample at different moments of time τ at $K_{CA} = K_{CB} = 1$, $K_{\lambda A} = 0,2$, $K_{\lambda B} = 5$; the center of the contact patch is at the point with the coordinate ξ : $0,2$ (a); $0,8$ (b); $3,2$ (c); $8,0$ (d).

Figures 4, 5 show temperature distributions at different ratios of thermal conductivity coefficients of layers A and B. As the thermal conductivity coefficient of layer A increases (Fig. 4), there is a faster heating of this layer in front of the roll, but the temperature value decreases. In the opposite situation (Fig. 5), the temperature in the adhesive layer changes much more slowly.

The presented examples do not allow us to unambiguously judge about the establishment of the heat propagation regime - stationary or quasi-stationary, and the structurization process. If a finite-size sample is considered, it is more convenient to study the behavior of the average integral value of the degree of structural transformations I_a and the average degree of structurization $\langle a \rangle$ over the thickness of the adhesion layer to determine the thermal regime:

$$I_a = \frac{1}{L} \int_{\Delta_B}^{1+\Delta_B} \left(\int_0^{\bar{L}} a(\xi, \eta) d\xi \right) d\eta, \quad \langle a \rangle = \int_{\Delta_B}^{1+\Delta_B} a(\xi, \eta) d\eta.$$

The stationary mode will correspond to practically constant maximum temperature and invariable average integral degree of structural transformations:

$$\frac{\theta_{\max}^{j+1} - \theta_{\max}^j}{\theta_{\max}^j} < 10^{-5}, \quad \frac{I_a^{j+1} - I_a^j}{I_a^j} < 10^{-5},$$

where j — layer number in time (that is, the numerators are the change in value in one time step).

The steady-state time is influenced by the model parameters. Figure 6 shows that over time both steady-state conditions are met with an accuracy of 10^{-5} , but differently for different sets of parameters. For example,

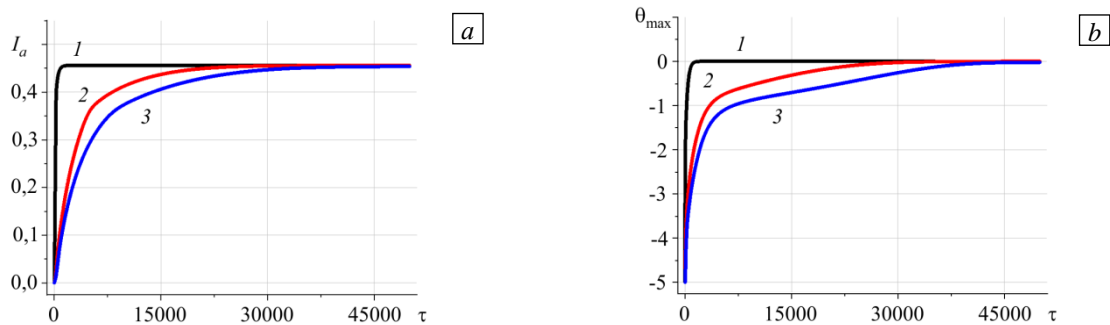


Fig. 6. Dependence of the average integral degree of structural transformations (a) and maximum temperature (b) on time at $\gamma = 0,5$, $\omega = 0,00048$, $\Pi = 1$, $\varepsilon = 0,87$, $\bar{a}_1 = \bar{a}_2 = 0$ and various values δ : 1 (curve 1); 50 (2); 100 (3)

at parameters $\gamma = 0,5$, $\omega = 0,00048$, $\Pi = 1$, $\varepsilon = 0,87$ the time of establishment of the stationary mode increases with growth δ : at $\delta = 1$ stationary mode occurs almost instantaneously, and at $\delta = 100$ stationary mode establishment time is 35000 (due to an increase in inhomogeneity in the temperature distribution).

Larger values δ correspond to greater thicknesses of the adhesive layer, which is identical to a decrease in the characteristic thermal scale equal to the size of the heating area that can be formed in a characteristic time.

The calculation results presented in Figures 2-6 were obtained without taking into account the effect on the kinetics of the sensitivity coefficients of the structurization reaction rates to the effective stresses. Figure 7a shows a significant effect of the $\bar{\alpha}_1$ and values $\bar{\alpha}_2$ on the degree of structurization: with increasing $\bar{\alpha}_2$ values, the average integral degree of structural transformations increases. A similar situation, as expected, is repeated in the character of the dependence of the average thickness distribution of the degree of transformation (Fig. 7b).

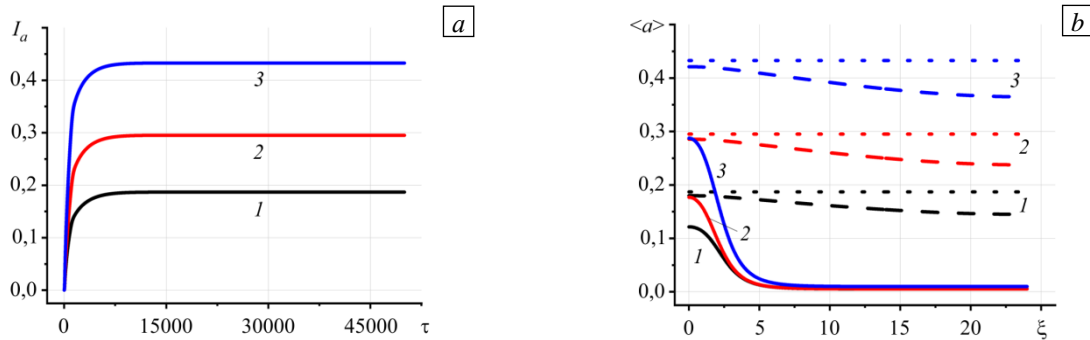


Fig. 7. Dependence of the average integral degree of structural transformations on time (a) at different moments of time τ : 100 (curve 1), 2500 (2), 15000 (3); distribution of the average degree of transformation along the thickness of the adhesive layer along the coordinate ξ (b) for different time moments τ : 100 (solid lines), 2500 (dashed lines), 15000 (dotted lines) at $\gamma = 1$, $\delta = 10$, $\omega = 0,00048$, $\Pi = 1$, $\varepsilon = 0,87$ and different combinations of values of dimensionless coefficients of sensitivity of reaction rates to stress $\bar{\alpha}$: $\bar{\alpha}_1 = 5, \bar{\alpha}_2 = 0,2$ (curves 1); $\bar{\alpha}_1 = 0,2, \bar{\alpha}_2 = 0,2$ (2); $\bar{\alpha}_1 = 0,2, \bar{\alpha}_2 = 5$ (3).

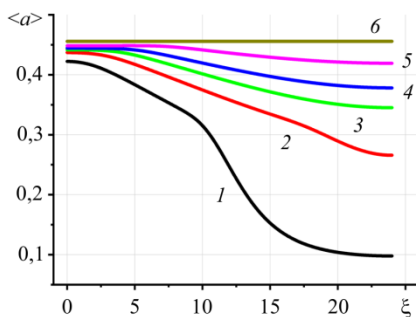


Fig. 8. Dependence of the average thickness of the adhesive layer on the degree of structurization at $\gamma = 0,5$, $\omega = 0,00048$, $\Pi = 1$, $\varepsilon = 0,87$, $\bar{\alpha}_1 = \bar{\alpha}_2 = 0$ for different time moments τ : 2500 (curve 1); 5000 (2); 7500 (3); 10000 (4); 15000 (5); 50000 (6).

The degree of structurization in the adhesive layer by the end of the lamination process is a function of all parameters. Thus, as the roll speed increases, the heat input decreases and, consequently, the average integral degree of structural transformation decreases. For the same reason, the time to the end of the technological process is shortened (Fig. 8). The layer is only partially structured, and the accumulated heat is not sufficient to increase the degree of structurization. The average integral degree of structural transformations reacts weakly to the growth of roll pressure (which can be explained by the expansion of the contact area and, consequently, by the growth of temperature in the adhesive layer), but the time for establishing the steady-state value becomes noticeably shorter I_a .

Examples of time establishment of steady-state value I_a dependencies on model parameters are shown in Figures 9-12. As the roll speed increases,

the upper layer of the sample warms up faster, but the heat may not reach the adhesion layer, which reduces the degree of structural transformations and, consequently, lengthens the steady-state time for I_a (see Figure

9). Figure 10 shows that an increase in roll pressure leads to a decrease in the steady-state time for I_a regardless of the value of δ .

In contrast to the technological characteristics, the change in the kinetic parameters ε and γ leads to a significant change in the value of the average integral degree of structural transformations by the time the quasi-stationary regime is established. An increase in ε entails an increase in ϕ_1 (see (14)) and, accordingly, an acceleration of the reverse structurization reaction, although it may start later; as a result, the average integral degree of structural transformations significantly decreases, as shown in Fig. 11a. The time for establishing the steady state value of I_a increases sharply.

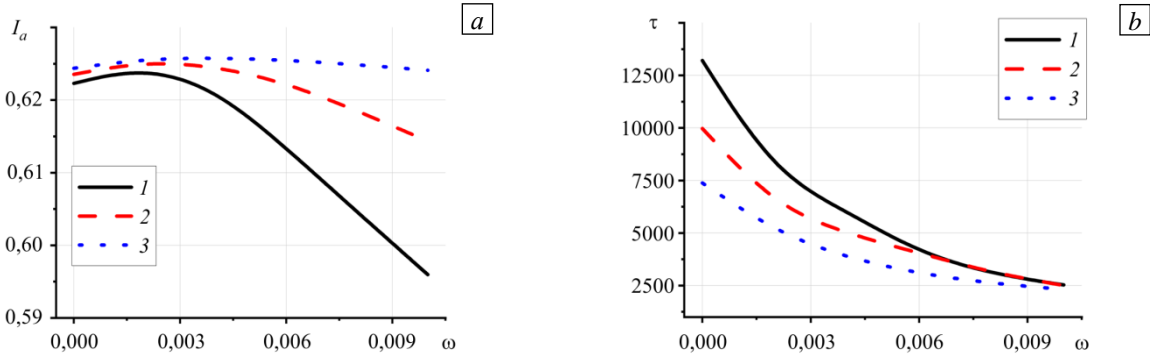


Fig. 9. Dependence of the average integral degree of structural transformations (a) and the time of establishment of the stationary value I_a (b) on the velocity of the roll at $\gamma = 1$, $\delta = 10$, $\varepsilon = 0.87$, $\bar{\alpha}_1 = \bar{\alpha}_2 = 0$ and different values of roll pressure Π : 0,1 (curve 1); 1 (2); 10 (3).

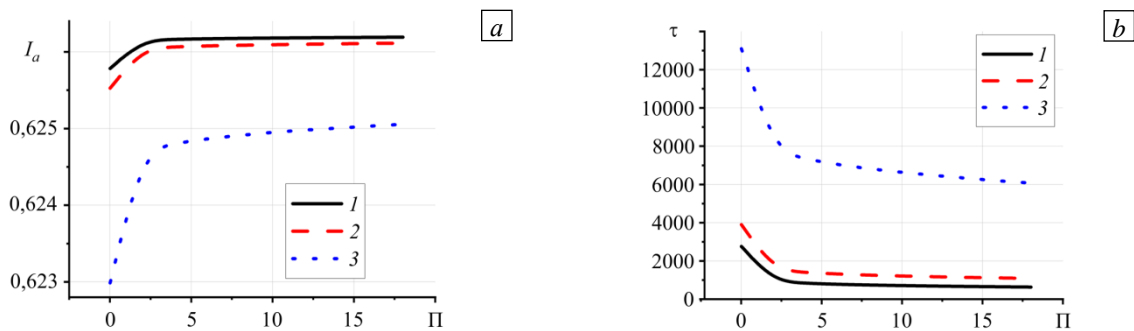


Fig. 10. Dependence of the average integral degree of structural transformations (a) and the time of establishment of the stationary value I_a (b) on the pressure of pressing the roll at $\gamma = 1$, $\omega = 0.00048$, $\varepsilon = 0.87$, $\bar{\alpha}_1 = \bar{\alpha}_2 = 0$ and different values δ : 0,5 (curve 1); 1 (2); 10 (3).

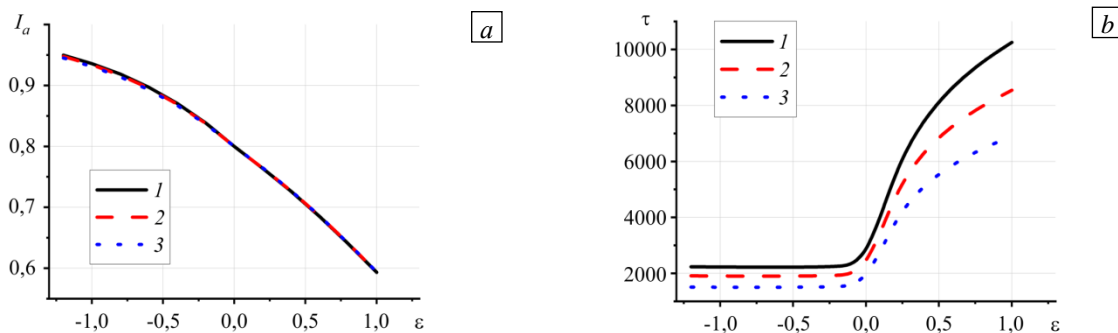


Fig. 11. Dependence of the average integral degree of structural transformations (a) and the time of establishment of the stationary value I_a (b) on ε at $\gamma = 1$, $\delta = 10$, $\omega = 0.00048$, $\bar{\alpha}_1 = \bar{\alpha}_2 = 0$ and different values of roll pressure Π : 0,1 (curve 1); 1 (2); 10 (3).

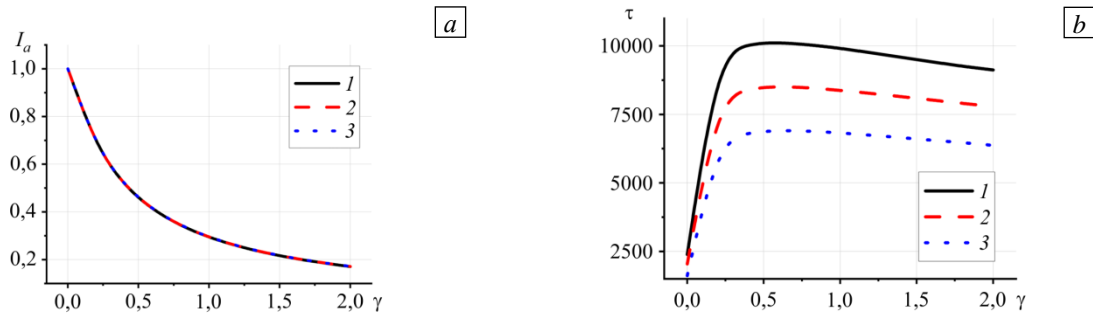


Fig. 12. Dependence on the kinetic properties of the average integral degree of structural transformations (a) and the time of establishment of the stationary value t_a (b) for $\delta = 1$, $\omega = 0,00048$, $\varepsilon = 0,87$, $\bar{\alpha}_1 = \bar{\alpha}_2 = 0$ and different values of roll pressure π : 0,1 (curve 1); 1 (2); 10 (3).

The value I_a significantly decreases as the parameter γ increases, while the time required establishing its stationary value sharply increases up to $\gamma = 0,45$ (see Fig. 12), which is again associated with the acceleration of the reverse structurization stage.

5. Conclusion

Thus, a thermo-physical model of the structurization process in the adhesive layer is proposed in the paper. The presence of direct and reverse stages in the structurization and the dependence of their rates on both temperature and stress leads to the fact that incomplete structurization is possible by the end of the roll motion. This indicates the necessity of selecting technological conditions for obtaining a given structure of the adhesion layer. The ratios of thermal physical properties of the inert layers and their geometrical sizes also affect the character of temperature distribution and the degree of structural transformations. At identical values I_a , attained at different sets of parameters, the termination end times of the structurization reaction may differ greatly. It has been shown that consideration of the forward and reverse stages, as well as the dependence of stage rates on temperature and stress, makes it possible to detect both the incompleteness of transformation, the presence of a quasi-stationary regime, and the effect of external controlling factors on the process.

We hope that the described regularities will prompt experimenters to control not only the mechanical properties of manufactured laminates, but also the values characterizing the process of object creation itself (time of start of transformations, size of area where structural transformations take place, time of structurization end and others), which is important for identification of the parameters included in the model.

Note that the role of individual parameters was not specifically studied in this work. And the fact that the sensitivity coefficients of stage velocities affect the value of the degree of structural transformations is demonstrated only by an example. The ratio of thermal conductivity coefficients of structured (λ_1) and unstructured (λ_2) materials $\kappa = \lambda_1/\lambda_2$ in calculations of the curves in all figures was fixed and equal to 0.1; that is, it was assumed that thermal conductivity coefficients after and before structurization differ by an order of magnitude. However, separate results not presented in the paper showed that the increase leads to a more homogeneous temperature distribution in the adhesive layer and accelerated heat outflow to the lower layer. Such findings will be of interest with respect to the specific materials for which the structurization kinetics will be studied.

There is information about the incompleteness of transformations in the structure and its dependence on the experimental conditions in the experimental works [27, 28]. The velocity value $\omega = 0,01$ means that during the time t_a the roll passes a distance 100 times less than the adhesive layer thickness. This can ensure its almost uniform heating over the thickness, but the presence of thermal conductivity and heat loss to the lower layer complicate the situation. The value $\delta = 50$ means that the thickness of the adhesive layer is 50 times greater than the distance by which the layer warms up due to thermal conductivity in the same time t_a . In the article, this parameter varied widely. An increase in the roll velocity requires, as in the experimental studies, an increase in its temperature in order to start the structurization process. If the scale ratios are changed, the dynamics of the process will obviously change. However, the criteria used in the article will be useful in any case.

Determining the kinetic parameters included in the model, as well as the nature of the dependence of the properties on the degree of structurization on the basis of computational experiment data, is the subject of further research. The proposed model can be improved by taking into account the temperature dependence of properties, as well as by explicitly formulating and solving the related problem of finding the stress-strain state of an object created by the LOM method.

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