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INVESTIGATION OF STRUCTURE FORMATION PROCESSES IN LIME-BASED RESTORATION COMPOSITES

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Abstract. The operational properties of composite materials are largely determined by the processes that occur at the beginning of their structure formation. Volumetric changes and plastic strength are considered as informative indicators reflecting the processes of structure formation and adaptation of lime restoration composites (plaster compositions) in the early stages of hardening. It is shown that these characteristics are sensitive to changes in the internal structure of the material, in particular to the coagulation of dispersed particles, the development of interphase contacts, and the formation of a spatially connected composite structure. The results demonstrate that volumetric changes and plastic strength reflect different but interrelated aspects of a single process–the formation of the internal structure of the composite and therefore can be considered as complementary indicators of structural evolution. The correlation between these indicators determines the ability of the restoration mortar to adapt to the deformation and moisture exchange characteristics of the porous limestone base without accumulating critical internal stresses. Based on experimental data, regression models and graphical dependencies were constructed, which clearly reflect the influence of composition on the nature of volumetric deformations and the kinetics of plastic strength growth. The obtained isosurfaces and time curves allow interpreting the change in properties through the dominance of certain mechanisms of structure formation at different stages of hardening. It has been shown that varying the composition of the composite allows for targeted control of the balance between deformability and the rate of increase in structural cohesion, which is a key factor in the formation of a compatible and stable contact zone. The results obtained confirm the possibility of using volumetric changes and plastic strength as a methodological basis for predicting the behavior of restoration mortars and adaptation processes in the "restoration mortar – base material" system and for designing plaster compositions focused on long-term compatibility and preservation of historic shell limestone buildings.

Keywords: structure formation, volumetric deformations, plastic strength, composite materials, adaptation, lime mortar, restoration composites.

Introduction. When carrying out restoration work on historic buildings made of shell limestone, an important stage is the design of plaster restoration composites. Along with the use of modern innovative materials for finishing and restoring shell limestone buildings, traditional lime mortars with fillers and additives remain relevant. The use of such composites ensures maximum compatibility in terms of vapor permeability and mechanical properties of the materials used with the base, which allows preserving the natural moisture exchange regime and maximum authenticity of the structure.

When researching lime composites with the aim of obtaining materials with the appropriate level of operational properties, dependencies of the "composition-technology-property" type are traditionally used, in which the main focus is on the quantitative content of individual components and the technology for obtaining materials. At the same time, as a rule, the internal organization of the material is not taken into account, the reasons for synergistic or antagonistic effects when combining components are not explained, and the empirical models obtained are poorly suited for analyzing the temporal evolution of properties. As a result, their predictive ability is limited, and the connection with the physical nature of the observed phenomena remains indirect. Overcoming these limitations requires a transition from a purely compositional description to a thorough analysis of the structure-forming processes occurring in the material, with the aim of further targeted control of the necessary operational properties. Thus, the study of the dynamic processes of structure formation in composite materials is a highly relevant task.

The properties that can give an idea of the processes of structure formation in the studied composites include volumetric deformations that occur during the hardening of restoration mortars and the plastic strength kinetics. These properties are particularly important for predicting the adaptive behavior of plaster mortars in the "plaster layer – shell limestone" system, as they ensure the coordination of composite deformations with the deformation response of the porous carbonate base and minimize internal stresses in the contact zone.

Thus, research and analysis of volumetric deformations and plastic strength in lime-based restoration composites is an important task for obtaining materials with the necessary set of operational properties and maximum compatibility with shell limestone.

Analysis of recent research and publications. When designing lime-based restoration composite compositions, the main task is to ensure maximum compatibility between the mortars and the base material [1, 2]. For porous carbonate rocks, such as shell limestone, compatibility is ensured by the consistency of both physicochemical and mechanical properties, as well as structural indicators and the formation of a mutually adapted "plaster layer – porous carbonate base" system [3-5].

The formation of the properties of composite building materials is inextricably linked to the processes of structure formation, which unfold over time and cover a wide range of spatial scales [6-8]. In binding systems and composites based on them, it is the structure that arises and evolves during the hardening process that determines the mechanical, deformation, and rheological characteristics of the material, as well as the patterns of their temporal change. At the same time, the structure of the material is not predetermined or static – it is the result of a complex non-equilibrium process of growth, restructuring, and interaction of structural elements, which includes coagulation and aggregation of particles, the formation of contact bonds, various interfaces and microcracks, the development of clusters, and the transition to a coherent, percolation network [6-9].

Significant characteristics are not so much the specific values of the parameters of a fixed structure at a given moment in time, but rather the ways in which they develop – that is, the processes of structure formation. It is the evolution of the structure, rather than its final state, that is more closely related to the general laws of physics, chemistry, and geometry of the processes occurring in composite materials [6, 9]. An essential aspect of such a structure-oriented approach is to consider structure formation processes not as the independent growth of individual subsystems, but as their mutual adaptation and restructuring. In this context, it is appropriate to use the concept of adaptation of composite components to each other [10], which manifests itself in the emergence of cooperative effects, changes in the kinetics of structure formation, and the formation of an effective structure whose properties cannot be reduced to the simple sum of the properties of individual components.

The integral-structural properties that determine the dynamics of structural processes include, for example, volumetric changes in the hardening composite and plastic strength. Plastic strength characterizes the emergence and development of the structural network, while volumetric changes reflect its compaction and reduction in free volume. Despite the difference in physical nature, both properties are determined by a single process of structure formation.

The objective processes that occur during the formation of the structure of filled composite materials cause a change in the volume of both local areas and the volume of the entire system as a whole. When filler particles are introduced into the matrix material of the binder, processes occur that lead to the formation of structural aggregates – clusters. Also, a layer appears on the surface of the fillers, the structure and properties of which differ from the properties of the composite in volume and depend to a large extent on the structure of the filler surface. As a result of such processes of forming the structure of building composites, density fluctuations occur, leading to volumetric deformations of the hardening composite mortar [11]. The uneven spatial distribution of structural elements causes local stresses, which in the early stages of hardening can partially relax due to plastic deformations, and with the subsequent increase in the rigidity of the system, are fixed in the form of shrinkage deformations. Thus, the nature and magnitude of volumetric changes are determined not only by the phase composition of the composite [12], but also by the kinetics of structure formation and the interaction between the matrix and the surface of the fillers.

The plastic strength of composite binder systems in the early stages of hardening is an integral characteristic that reflects the formation of a load-bearing structural network capable of transferring stress in the mode of irreversible deformation [13-15]. Unlike fracture strength, which is determined by the ultimate stability of the formed contacts, plastic strength is formed in the process of gradual structure formation and is sensitive to the topology, connectivity, and heterogeneity of the growing network.

Physical plastic deformation in composites as systems corresponds to the collective rearrangement of contacts between structural elements – binder particles, hydration products, additive particles, filler grains, and their aggregates. Therefore, plastic strength is not an instantaneous value, but develops over time as a result of a relaxation process associated with the growth and rearrangement of the cluster-network system.

The change in the plastic strength of plaster composites over time reflects a complex multistage process of internal material structure formation, which includes the coagulation of dispersed particles, the formation of contacts between solid phases, and the gradual transition of the system from a viscoplastic to a shear-deformation state [13, 14, 16, 17]. In the early stages of hardening, plastic strength is determined mainly by physical interparticle interactions caused by van der Waals forces, electrostatic and capillary effects, which form a coagulation structure with reversible contacts. In this case, shear resistance arises from the cumulative effect of a large number of weak bonds, and plastic strength can be described within the framework of the classical Bingham rheological model [17, 18], where the yield stress τ is identified with the plastic strength of the material (1):

$$\tau = R_{pl} + \eta\gamma, \tag{1}$$

where R_{pl} – plastic strength (ultimate shear stress), Pa; η – plastic viscosity, Pa·s; γ – shear rate, s^{-1}.

As the material hardens, the R_{pl} value increases, reflecting an increase in the degree of structural cohesion of the system.

Further development of plastic strength is associated with the transition from a purely coagulation structure to a coagulation-crystallization structure, when, along with physical contacts, more stable chemical and physicochemical bonds appear, caused by the processes of hydration of binding components, sorption interactions of dispersed particles, and gradual crystallization of hardening products. From the point of view of physically-chemical mechanics [16], the increase in plastic strength during this period may be associated with an increase in the number of contacts between particles and their average strength, which is generally described by the relationship (2):

$$R_{pl} = k \cdot n \cdot F_c, \tag{2}$$

where n – the number of effective contacts per unit volume, m^{-3}; F_c – the average interparticle interaction force, N; k – the geometric coefficient.

Thus, plastic strength increases both as a result of structure densification (increase in n) and as a result of qualitative evolution of the contacts themselves (increase in F_c).

In terms of time, plastic strength is not a constant material characteristic, but rather an integral parameter that reflects the current state of the material's structure formation. Its kinetics are determined not only by the speed of chemical or physicochemical reactions, but also by the consistency of geometric and mechanical connections between the components of the composition, the degree of formation of a spatially connected framework, and the ability of the system to redistribute deformations [19, 20]. That is why plastic strength is a sensitive indicator of the early hardening of plaster composites and allows a quantitative assessment of the transition of the material from a rheologically controlled state to a structurally stabilized one, which is of fundamental importance for the technological and operational reliability of plaster and restoration systems.

Thus, by analyzing the volumetric changes and plastic strength of restoration lime composites, it is possible not only to quantitatively assess the intensity of early structure formation processes, but also to predict the adaptive behavior of the material in the "plaster layer – stone base" system. A combined consideration of these two characteristics allows us to determine the extent to which the formed structure of the composite is capable of coordinating its own deformations with the deformation response of the shell limestone base, which is characterized by increased porosity, anisotropy, and sensitivity to moisture exchange processes.

Volumetric deformations occurring in the early stages of hardening determine the level of internal stresses in the plaster layer and in the contact zone with the base. Excessive volume changes in conditions where the structure is not yet sufficiently formed can lead to local stress concentration, disruption of adhesive bonds, and the initiation of microcracks in the surface layers of shell limestone. At the same time, the development of plastic strength characterizes the material's ability to relax these stresses through creep and redistribution of deformations without losing the integrity of the layer. That is why plastic strength at a fixed moment of early hardening is an indicator of whether the system is transitioning from a deformation-unstable state to an adapted, structurally bound mode of operation.

Thus, a comprehensive analysis of volumetric deformations and plastic strength allows us to predict not only the early kinetics of hardening, but also the long-term operational behavior of the restoration material in contact with historic masonry. This is fundamentally important when designing compositions for the restoration and renovation of shell limestone buildings, as it ensures the formation of an adaptive, compatible, and structurally stable "plaster – base" system capable of functioning without accumulating destructive stresses during operation.

Purpose of the work is to research into the kinetics of volumetric deformations and plastic strength of lime-based restoration composites in order to identify patterns of early structure formation and adaptation and to predict the compatibility of composite materials with porous limestone bases.

Materials and methods of research. The composite material under study is intended for use as a plastering mortar in conservation and restoration works. The main components of the studied mortar are air lime, Portland cement, clay additive (kaolin clay), quartz sand, and crushed limestone (specific surface area 3000 cm^2/g) as a structurally compatible filler. The quantitative ratios and levels of variation of the composition factors in the planned experiment according to a 15-run experimental design are shown in Table 1.

Table 1 – Factors of the planned experiment and their levels of variation

№	Factors	Coded values			Values, %		
		Min	Center	Max	Min	Center	Max
X_1	The content of cement in the total mixture of binders	-1	0	1	5 %	10%	15 %
X_2	The content of clay in the total mixture of binders	-1	0	1	5 %	7,5%	10 %
X_3	The content of ground limestone in the filler	-1	0	1	0 %	37,5%	75 %

We monitored the volumetric deformations of hardening compositions and changes in plastic strength over time.

Volumetric deformations were determined using a specially designed device, the diagram of which is shown in Fig. 1.

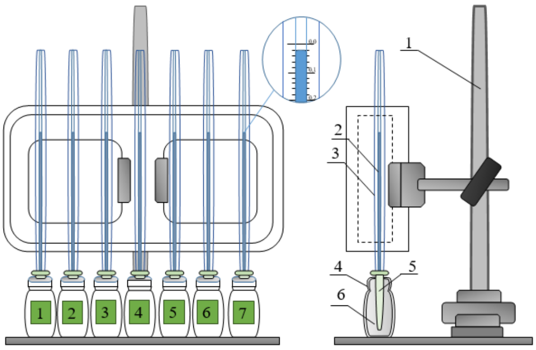


Fig. 1. Device for determining volume changes in hardening mortars:
1 – stand; 2 – colored liquid; 3 – pipette; 4 – container for mortar; 5 – rubber tip; 6 – hardening mortar

Volumetric changes were calculated using the formula (3):

$$\Delta V = \frac{\Delta V_{pipette}}{V_{mortar}} \cdot 100\%, \tag{3}$$

where V_{mortar} – the volume of the mortar, mm^3; $\Delta V_{pipette}$ – the difference in volume values of the colored liquid, mm^3 (4):

$$\Delta V_{pipette} = V_{pipette_{n+1}} - V_{pipette_n}. \quad (4)$$

The plastic strength was determined using the Rebinder method with a conical plastometer, Fig. 2.

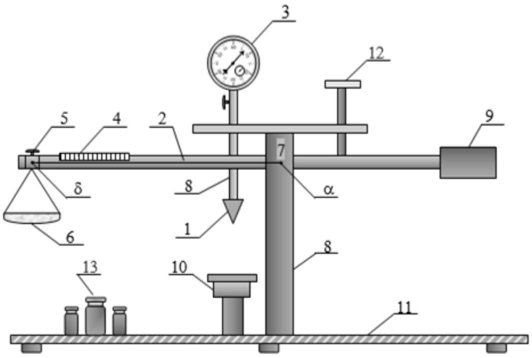


Fig. 2. Rebinder's conical plastometer: 1 – metal cone; 2 – lever (rocker arm); 3 – indicator; 4 – ruler; 5 – clamp; 6 – tray; 7 – lever suspension point; 8 – stand; 9 – counterweight; 10 – movable table; 11 – base plate; 12 – bolt; 13 – set of weights

Research results. Let us consider the processes of structure formation in the studied lime composites that contribute to the formation of volumetric deformations. Volumetric changes in hardening mortars in the studied time interval are characterized by monotonic negative deformation and therefore can be further considered as contraction.

Volumetric changes (contraction) in a hardening plaster restoration compound based on lime with a small addition of cement, clay, and a combined filler (sand + crushed shell limestone) are the result of the combined action of physicochemical and structural-mechanical processes occurring at different scale levels – from the nanopores of the binder matrix to the structure of the intermediate scale level of the filled composite. Contraction is formed as an integral effect of the restructuring of the solid phase, pore space, and interphase interfaces and microcracks during hardening and drying. In the early stages of structure formation, the formation of a spatial network of contacts between the particles of the lime binder and the products of cement hydration plays a decisive role.

Hydrated lime $Ca(OH)_2$ forms a dispersed colloidal system in which the initial contraction is caused by particle coagulation and compaction of flocculated aggregates. This process can be described by the decrease in gel volume V_g over time t (5):

$$\varepsilon_g(t) = \frac{V_g(t) - V_{g0}}{V_{g0}} < 0, \quad (5)$$

where V_g – the initial volume of the gel phase.

The addition of a small amount of cement leads to the formation of calcium hydrosilicates (C-S-H), which have significantly higher specific surface energy and self-compacting properties. The chemical shrinkage of the cement component can be expressed by the difference in the molar volumes of the reactants and hydration products (6):

$$\varepsilon_{ch} = \frac{\sum_i \nu_i V_i^{\text{prod}} - \sum_j \nu_j V_j^{\text{react}}}{\sum_j \nu_j V_j^{\text{react}}}, \quad (6)$$

where ν_i, ν_j – stoichiometric coefficients; V_i^{prod}, V_j^{react} – molar volumes of the corresponding phases.

The further drying and carbonization of lime is accompanied by the development of capillary stresses P_c in pores with radius r, the magnitude of which is described by the Kelvin–Laplace equation (7):

$$P_c = -\frac{2\gamma\cos\theta}{r}, \quad (7)$$

where γ – the surface tension of a porous fluid; θ – contact angle of wetting.

The integration of capillary stresses according to the distribution of pores by size causes macroscopic shrinkage deformation (8):

$$\varepsilon_{cap} = \frac{1}{K_{eff}} \int_{r_{min}}^{r_{max}} P_c(r) \cdot f(r), dr, \tag{8}$$

where K_{eff} – the effective volumetric modulus; $f(r)$ – function of pore size distribution by radius.

Clay, introduced in small quantities, performs a dual role. On the one hand, its lamellar particles adsorb water and increase the initial volume of the gel phase; on the other hand, during drying, interlayer convergence occurs, accompanied by additional contraction. This contribution can be described as sorption-induced deformation (9):

$$\varepsilon_{cl} = k_{cl} \cdot \Delta w, \tag{9}$$

where Δw – the change in the content of bound water; k_{cl} – the sorption deformation coefficient of clay.

The combined filler forms the structure of the intermediate scale composite and determines the nature of the interphase transition zone. Quartz sand is inert and limits volume reduction due to its mechanical framework, while porous shell limestone is partially involved in moisture exchange and locally enhances capillary effects. The total shrinkage of the filled composite can be approximated by the following model, taking into account the stiffness of the phases (10):

$$\varepsilon_{tot} = \varepsilon_m \frac{K_m}{K_m + K_f \phi_f}, \tag{10}$$

where ε_m – the matrix shrinkage; K_m i K_f – the bulk modulus of the matrix and filler; ϕ_f – the volumetric fraction of filler.

An important mechanism is the formation of density fluctuations in the transition zone "binding agent – filler". Here, due to differences in the rate of hydration, carbonization, and drying, local deformation gradients arise, which, when integrated over volume, cause macroscopic contraction and internal stresses. It is these stresses that determine the tendency of the restoration composition to microcracking or, conversely, to relaxation due to the creep of the lime matrix.

The effect of ground shell limestone on the volume changes of hardening lime-cement composites is multicomponent and depends on its dispersibility, porosity, and mechanical compatibility with the lime matrix. On the one hand, the introduction of finely dispersed limestone reduces the effective proportion of the binder phase per unit volume of the composite, which leads to a decrease in chemical and gel shrinkage (11):

$$\varepsilon_{eff} = \varepsilon_m (1 - \phi_{lim}), \tag{11}$$

where ϕ_{lim} – volume fraction of ground shell limestone.

On the other hand, the high specific surface area and open porosity of shell limestone particles contribute to moisture redistribution in the system. Some of the water accumulates in the pores of the filler and is gradually released during the hardening process, which reduces moisture gradients and the intensity of capillary shrinkage. This leads to a reduction in maximum capillary stresses. At the same time, the microporous structure of ground shell limestone is partially involved in sorption-type deformations, but these deformations are dispersed and do not form local stress concentrators. At the intermediate scale, shell limestone acts as a "deformation damper", reducing the total internal stresses σ_{red} in the transition zone "binding agent – filler" (12):

$$\sigma_{red} = \sigma_0 \left(1 - \frac{\phi_{lim}}{E_{eff}} \right), \tag{12}$$

where σ_0 – the initial stresses in the matrix; E_{eff} – the effective modulus of the composite, reduced due to the porous filler.

Thus, volumetric deformations of hardening lime-cement restoration composition are the result of the formation of structural aggregates of dispersed particles, chemical shrinkage of hydration and carbonation reactions, capillary shrinkage during water removal, sorption deformations of clay, and structural-mechanical interaction with a combined filler. An adequate description of volumetric deformations is possible only on the basis of multilevel models of structure formation.

The formation of plastic strength in lime-based plaster restoration composites with a small addition of cement, clay, and combined filler reflects the formation of the internal load-bearing structure of the material in the early stages of hardening. In terms of time, plastic strength characterizes the transition of the system from a viscous-plastic, rheologically active state to a shear-deformation state, in which the ability to perceive and transmit mechanical stresses without loss of integrity is formed.

The main binder in the system is lime, which forms the initial dispersed medium with high water retention capacity and ensures the prolonged plastic state of the composition. In the first hours after mixing, the plastic strength of such mortars is determined mainly by the coagulation structure that arises as a result of physical contacts between particles of calcium hydroxide, clay minerals, and finely dispersed filler components. That is, in the initial stages of hardening, the lime binder forms a dispersed colloidal system. The increase in plastic strength at this stage can be described by a generalized kinetic relationship (13):

$$R_{pl}(t) = R_{pl,0} + A \cdot t^{n}, \tag{13}$$

where $R_{pl,0}$ – the initial value of plastic strength; A – the structural-kinetic coefficient; $n<1$ – a parameter that reflects the coagulation nature of structure formation.

The introduction of cement in an amount of 5-15% of the total binder component mass causes the activation of early structure formation processes due to the hydration of clinker minerals, primarily C_3A, and surface hydration of C_3S. The products of these reactions in the early stages are local in nature and do not form a solid rigid framework, but they significantly increase the number and strength of contacts between the solid phases. This manifests itself in an increase in plastic strength already in the first hours of hardening, while maintaining sufficient deformability of the system. Thus, cement in the studied compositions acts as a structure-forming additive that controls the rate of transition of the material from a viscous-plastic to a shear-deformation state.

The introduction of a small amount of cement leads to the formation of local rigid nodes due to early hydration products, which reduce contact reversibility and increase the effective shear modulus of the structure. In this case, plastic strength correlates with an increase in contact density (14):

$$R_{pl} = k \cdot z \cdot f_{\mathrm{cont}}, \tag{14}$$

where z – the coordination number of contacts; f_{cont} – the average force of interparticle interaction.

Clay, added in an amount of 5-10% of the binder mass, significantly affects the formation of plastic strength due to its high specific surface area, sorption capacity, and interlayer interactions. Clay particles retain water in an adsorbed state, reduce its migration rate, and contribute to the stabilization of the coagulation structure. This can ensure a smoother and more controlled increase in plastic strength over time, reducing the risk of sudden deformations and local failures in the plastic state. At the same time, excessive clay content can lead to an increase in water demand and a certain slowdown in the formation of a structurally bound framework, which is reflected in a shift of the maximum rate of plastic strength growth to later stages.

Ground shell limestone with a specific surface area of approximately 3000 m^2/kg in restoration plaster composites not only acts as an inert filler, but is also an active structure-forming component in the early stages of hardening. Its effect on plastic strength is realized mainly through physical-mechanical and geometric mechanisms, rather than through chemical activity in the classical sense. Due to their high dispersibility and developed surface, ground limestone particles are actively involved in the formation of the coagulation structure of the lime matrix. In the early stages of hardening, they act as centers of adsorption of $Ca(OH)_2$ colloidal particles, contributing to an increase in the density of contacts in the structural network. In simplified terms, the contribution of ground limestone to plastic strength can be described by an increase in the coordination number of contacts (15):

$$R_{pl,lim} = R_{pl,0} + k \cdot \Delta z_{lim} \cdot f_{\mathrm{cont}}, \tag{15}$$

where Δz_{lim} – the increase in the number of contacts caused by the presence of dispersed limestone filler.

The porous microstructure of shell limestone ensures partial absorption of free water from the liquid phase, which leads to a local decrease in water content in the interparticle space. This contributes to early compaction of the structure and faster growth of plastic strength without the formation of a rigid crystalline framework. Thus, the growth of R_{pl} occurs gradually, preserving the rheological "performance" of the mortar, which is fundamentally important for restoration plastering works.

In addition, the chemical affinity of the calcite surface of ground limestone with lime carbonation products reduces the energy heterogeneity of the interphase zone and ensures a smooth transition from a coagulation to a contact-skeleton structure. As a result, plastic strength is formed evenly, without sharp jumps, which reduces the risk of early structural cracking.

Thus, the plastic strength of the studied lime-cement-clay compositions is formed as an integral result of coagulation processes, early cement hydration, sorption properties of clay, and the structure-forming effect of finely dispersed carbonate filler. Its temporal development reflects a gradual increase in the degree of structural cohesion of the material and can be considered as a quantitative indicator of the mortar's readiness to perceive early deformations, which is fundamentally important for the use of such compositions in restoration work on porous limestone bases.

The plastic strength curves can be interpreted as reflecting the scenario of composite structure formation, in which different components do not simply add their properties, but interact, changing the pace and nature of the formation of the load-bearing network. This interpretation allows us to link the experimental plastic strength curves with the composition of the material and the physical content of the structure formation processes.

Based on the results of the experimental research and determination of volumetric changes and plastic strength over time during the hardening of restorative composites, experimental statistical models were obtained in the form of second-order regression equations (16), (17):

$$\Delta V = 0,221 + 0,070x_1 - 0,026x_2 + 0,028x_3 + 0,025x_1x_2 + 0,027x_3^2\,, \tag{16}$$

$$R_{pl} = 95,2 + 19,9x_1 + 5,5x_2 + 12,4x_3 - 7,6x_1^2 - 4,6x_2^2 - 7,1x_3^2 + 0,9x_1x_2 + 1,9x_1x_3\,. \tag{17}$$

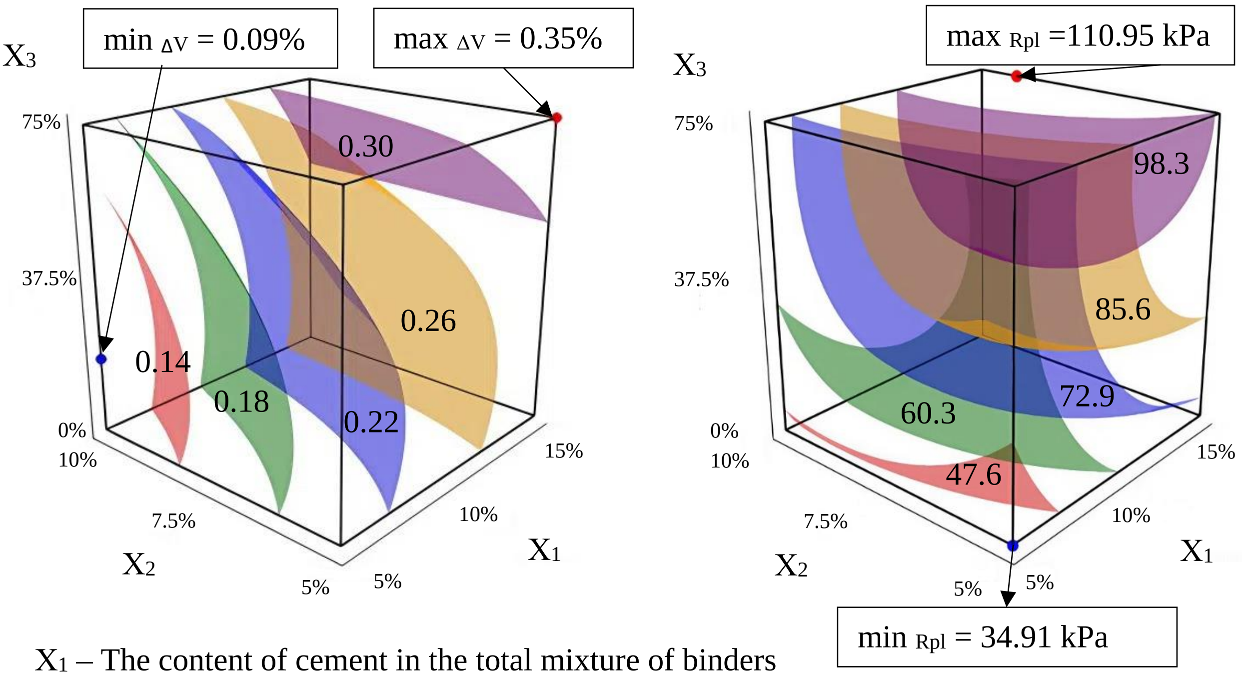
It should be noted that the fixed values of volumetric changes on which the model is based correspond to the maximum contraction of composite mortars, and the values of plastic strength used in modeling corresponded to 10 hours from the start of hardening. This time interval for determining the fixed value of plastic strength is due to the fact that 10 hours is the moment when plastic shrinkage is almost complete, the structure is already capable of perceiving its own deformations, transferring stress to the base (important for plaster mortar), not collapsing during drying, but the material has not yet become brittle. It is during this period that the "plaster- shell limestone" contact zone is formed, which determines the durability of the restoration system as a whole. In other words, this is the optimal balance between adhesion, deformability, and crack resistance.

The graphical representation of the constructed models is shown in Fig. 3.

The regression models of volumetric deformations and plastic strength obtained reflect different but interrelated aspects of the structure formation process of lime-cement-clay plaster composites.

The volumetric deformation model has a relatively simple structure and is described by linear effects of factors and a limited number of nonlinear terms. A positive coefficient for cement content (b_1 = +0.070) indicates that an increase in the cement fraction intensifies the total contraction, which is a physically justified consequence of the chemical shrinkage of hydration products and an increase in capillary stresses in the early stages of hardening. The negative coefficient for clay (b_2 = –0.026) indicates its partial compensatory role due to its sorption capacity and increased deformability of the coagulation structure. The positive contribution of ground shell limestone (b_3 = +0.028) and the quadratic term $b_{33}>0$ reflect its participation in moisture exchange and the development of capillary shrinkage due to its porous structure, as well as an increase in shrinkage at high concentrations. The interaction of cement and clay (b_{12} = +0.025) confirms that their joint presence leads to a more intensive restructuring of the microstructure than the action of each component separately.

The plastic strength model is nonlinear, which corresponds to the nature of this property as an integral characteristic of the degree of structural cohesion of the material. Positive linear coefficients for cement (b_1 = +19.9), clay (b_2 = +5.5), and ground shell limestone (b_2 = +12.4) indicate their contribution to the formation of the load-bearing coagulation-frame structure. At the same time, negative quadratic terms (b_{11}, b_{22}, $b_{33}<0$) indicate the presence of composition optimums: an excessive amount of any of the components leads to structural oversaturation, disruption of contacts, or an increase in internal stresses, which limits further growth in plastic strength. Positive cement-clay and cement-shell limestone interaction coefficients confirm the synergistic nature of these combinations in the formation of early strength.



X_1 – The content of cement in the total mixture of binders
X_2 – The content of clay in the total mixture of binders
X_3 – The content of ground limestone in the filler

Fig. 3. Isosurfaces of models:
a – volumetric deformations (ΔV, %); b – plastic strength (Rpl, kPa)

A qualitative comparison of the two models shows that they describe different aspects of the same process. Volumetric deformations mainly reflect the early restructuring of the pore space and gel phase before the formation of the load-bearing framework, while plastic strength characterizes the ability of the formed structure to resist shear. Therefore, common factors (primarily cement and ground shell limestone) can increase both shrinkage and plastic strength, but not because of a direct causal relationship between these properties, but because of their dependence on the intensity of structure formation.

For mortars of two compositions, graphs of changes in volumetric deformations and increases in plastic strength were constructed (Figs. 4, 5). Mortar No. 3 has a composition (5% cement of the total mass of the binder, 10% clay of the total mass of the binder, quartz sand as a filler without the addition of ground limestone) that corresponds to the minimum contraction. Mortar No. 8 has a composition (15% cement of the total mass of the binder, 10% clay of the total mass of the binder, complex filler – 25% sand and 75% ground limestone), which corresponds to the maximum plastic strength.

The maximum contraction rate is observed within 1–3 hours from the start of hardening, since during this period the system does not yet have a formed load-bearing framework and is in a rheologically active, "living" state. The main deformations develop under conditions of minimal internal resistance of the structure, when volumetric changes are determined by physicochemical processes of water redistribution and compaction of the dispersed system due to the formation of structural aggregates. Capillary or plastic shrinkage, which occurs due to the formation of menisci in macro- and mesopores and the appearance of capillary pressure, plays a significant role at this time. With relatively large pore radii and the absence of a rigid structural grid, capillary pressure is not compensated by the internal resistance of the material, so deformations develop at maximum speed. At the same time, intensive compaction of the coagulation structure occurs: contacts between particles are physical and reversible, particles are easily displaced, and so-called self-packing of the system occurs, leading to a sharp decrease in volume at low values of plastic strength. At the same time, water binding processes begin – surface hydration of cement minerals, sorption and interlayer interactions in clay, dissolution of calcium hydroxide in lime components. However, these processes do not yet form a single spatially connected framework, but are local and unconnected in nature. It is the combination of intense capillary phenomena, coagulation compaction, the formation of aggregates of dispersed particles, and the absence of a supporting structure that causes the maximum shrinkage rate to occur in the early interval of 1-3 hours of hardening.

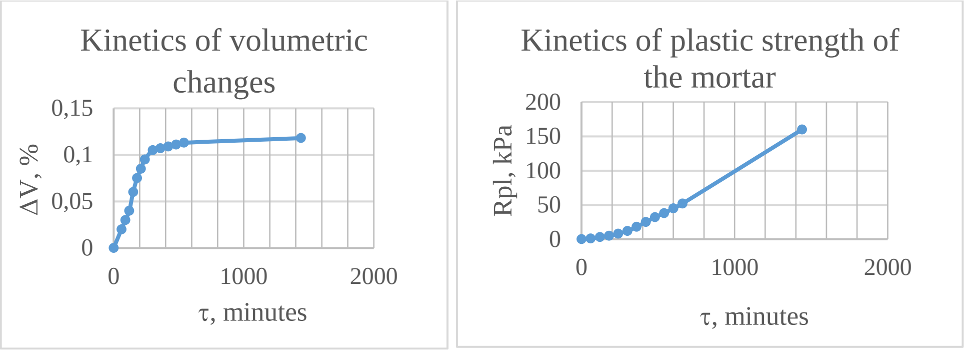


Fig. 4. Kinetics of volume changes and plastic strength of mortar No. 3

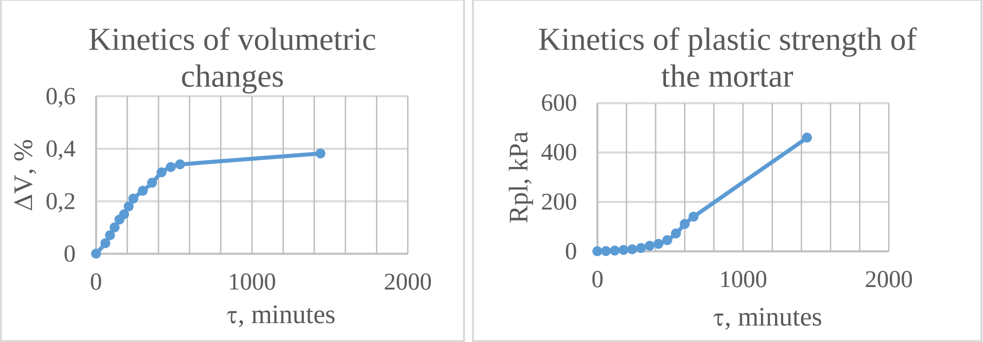


Fig. 5. Kinetics of volume changes and plastic strength of mortar No. 8

The obtained kinetics of plastic strength growth has a characteristic three-stage form, reflecting a sequential transition from a rheologically controlled state to the formation of a spatially bound structural framework. The initial period (up to 2-3 hours) is characterized by low plastic strength values with intense volume changes, while the subsequent increase in R_{pl} is accompanied by stabilization of shrinkage. This confirms the feasibility of using plastic strength as an indicator of early structure formation in lime-cement-clay compositions.

The second period (3–10 hours) is marked by accelerated growth of R_{pl}, indicating a transition from a coagulation structure to a contact-scaffold structure. At this time, the intensity of shrinkage decreases significantly, as the growing internal framework begins to resist capillary stresses. This is a critical period for restoration mortars, as the balance between structural compaction and the ability to relax internal stresses determines the further crack resistance of the material.

The third period (over 10 hours) is characterized by stabilization of R_{pl} growth rates and virtually complete shrinkage. The structure acquires the properties of a frame system with predominantly irreversible contacts, and further strength growth occurs due to chemical processes – cement hydration and slow lime carbonation.

Analysis of the graphs shows that mortar 3 with minimal contraction is characterized by a smoother but slower increase in R_{pl}, while mortar 8 demonstrates more intense early strength with slightly increased shrinkage. This confirms the possibility of purposeful regulation of the balance between deformability and structural cohesion through optimization of the content of cement, clay, and ground shell limestone.

Conclusions. It has been established that volumetric deformations and plastic strength of lime-cement-clay restoration composites are interrelated but not functionally dependent characteristics of the early structure formation process. Maximum contraction of up to 0.35% occurs in the first 24 hours of hardening, with a critical interval of 1-3 hours corresponding to the coagulation state of the system without a formed load-bearing framework. Plastic strength formation occurs in a three-stage process with a transition to a framework state after 10 hours, which corresponds to the completion of plastic shrinkage. It has been shown that the introduction of ground shell limestone reduces the intensity of volumetric deformations by 15–20% and increases early plastic strength by 1.5–1.8 times due to the compaction of the structure and the formation of additional coagulation contacts. The obtained second-order regression models enable the prediction of volumetric changes and plastic strength of composite materials as a function of composition and facilitate the targeted design of compatible restoration mortars for porous limestone substrates.

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ДОСЛІДЖЕННЯ ПРОЦЕСІВ СТРУКТУРОУТВОРЕННЯ РЕСТАВРАЦІЙНИХ КОМПОЗИТІВ НА ОСНОВІ ВАПНА

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Анотація. Експлуатаційні властивості композиційних матеріалів значною мірою обумовлені процесами, що протікають на початку формування їх структури. Об’ємні зміни та пластична міцність розглядаються як інформативні показники, що відображають процеси структуроутворення та адаптації вапняних реставраційних композитів (штукатурних складів) на ранніх стадіях твердіння. Показано, що зазначені характеристики чутливо реагують на перебудову внутрішньої структури матеріалу, зокрема на коагуляцію дисперсних частинок, розвиток міжфазних контактів і формування просторово зв’язаної структури композиту. Обґрунтовано, що об’ємні зміни та пластична міцність відображають різні, але взаємопов’язані аспекти одного процесу – становлення внутрішньої структури композиту, і тому можуть розглядатися як взаємодоповнювальні індикатори структурної еволюції. Узгодженість цих показників визначає здатність реставраційного розчину адаптуватися до деформаційних і вологообмінних особливостей пористої вапнякової основи, що реставрується, без накопичення критичних внутрішніх напружень. На основі експериментальних даних побудовано регресійні моделі та графічні залежності, які наочно відображають вплив складу на характер об’ємних деформацій і кінетику наростання пластичної міцності. Отримані за результатами дослідження ізоповерхні та часові криві дозволяють інтерпретувати зміну властивостей через домінування тих чи інших механізмів структуроутворення на різних етапах твердіння. Показано, що варіювання складу композиту дозволяє цілеспрямовано керувати балансом між деформативністю та швидкістю наростання структурної зв’язаності, що є ключовим чинником формування сумісної та стабільної контактної зони. Отримані результати підтверджують можливість використання об’ємних змін і пластичної міцності як методичної основи для прогнозування поведінки реставраційних розчинів, адаптаційних процесів у системі «реставраційний розчин – матеріал основи» та для проєктування штукатурних композицій, орієнтованих на довготривалу сумісність і збереження історичних будівель з вапняку-черепашнику.

Ключові слова: реставраційні композити, формування структури, об'ємні деформації, пластична міцність, адаптація, композиційні системи, розчини на основі вапна.

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