UDC 661.728:615.46:668.5
DOI: 10.15587/1729-4061.2025.341368

DEVELOPMENT OF THE COMPOSITION AND MODELING OF THE PROPERTIES OF THE BIOPOLYMER SYSTEM FOR GEL COSMETIC PRODUCTS WITH LONG-LASTING ACTION

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The object of the study is the properties of a biopolymer system for prolonged delivery of cosmeceutical active ingredients based on natural polysaccharides (sodium hyaluronate and chitosan).

The work solves the problem of the lack of a methodology for predicting the properties of a biopolymer system at the design stage. This methodology should link its component composition with functional and structural-mechanical characteristics. The optimization criterion should be achieving a controlled release profile of the hydrophilic active (dexpanthenol) without an initial "explosive" effect. It was established that the optimal ratio is 2.5% sodium hyaluronate and 1.2% chitosan, which ensures the release of 30–35% of dexpanthenol in 6 hours and 60–65% – in 24 hours. Structural and mechanical analysis confirmed the formation of a stable gel with a viscosity of 9800 ± 250 mPa·s and a storage modulus of 325 Pa. This is explained by the formation of a dense polyelectrolyte network due to electrostatic interactions between the anionic groups of sodium hyaluronate.

The results of the study of the biopolymer system include the development of approximation models. These models predict the release profile of dexpanthenol based on the concentration of polymers and evaluate the structural and mechanical properties of the biopolymer system. The results obtained can be used in the cosmeceutical industry to create gel products with prolonged action. The effectiveness of the system was confirmed in vitro in phosphate buffer solution (pH 7.4; 37°C). The criterion for effectiveness was the achievement of a controlled release profile: 30–35% of the active substance in 6 hours and 60–65% in 24 hours, which provides a long-term effect without an initial "explosive" effect

Keywords: biopolymer system, prolonged delivery, release kinetics, structural and mechanical properties, polyelectrolyte complex

Received 16.07.2025
Received in revised form 23.09.2025
Accepted date 03.10.2025
Published date 31.10.2025

How to Cite: *Putyatin, B., Bliznjuk, O., Panasenko, V., Belinska, A., Svishchova, Y., Zolotukhina, O., Ashtaeva, N., Masalitina, N., Zabrodina, I., Kovalova, T. (2025). Development of the composition and modeling of the properties of the biopolymer system for gel cosmetic products with long-lasting action. Eastern-European Journal of Enterprise Technologies, 5 (6 (137)), 6–13. https://doi.org/10.15587/1729-4061.2025.341368*

1. Introduction

The global cosmeceutical market is showing steady growth, driven by the demand for effective products with proven bioavailability and prolonged action. According to forecasts, its volume will reach 85.6 billion USD by 2030 [1], which actualizes the development of new delivery systems for active ingredients. Biopolymers of natural origin, in particular hyaluronic acid, chitosan and alginates, are a promising basis for such systems due to the combination of biocompat-

ibility, biodegradability and matrix-forming ability [2]. However, as researchers note, only 23% of the developed polymer systems demonstrate a reproducible release profile *in vivo*, which is associated with the difficulty of predicting the interaction of "composition-structure-function" [3]. A key problem that limits the widespread use of biopolymer matrices is the lack of quantitative models that link the physicochemical parameters of the system with its functional characteristics. For example, a study [4] showed that a variation in polymer concentration of only 0.5% can lead to a change in the release rate of the active ingredient by 35–40%, but did not propose a mechanistic model to explain this phenomenon. Other works [5, 6] state the problem of instability of finished products: up to 30% of emulsion systems based on biopolymers demonstrate stratification during the first 30 days of storage.

Thus, the need to develop cosmeceuticals with predictable properties indicates the relevance of research aimed at modeling the composition of biopolymer systems.

2. Literature review and problem statement

In the study [7], cellulose nanocrystals were successfully used as emulsifiers and thickeners in sunscreen emulsions, demonstrating good stability and performance characteristics comparable to commercial analogues. However, the work left unresolved the issue of the influence of the multicomponent composition of real cosmeceutical formulations, in particular the presence of salts, metal ions and other active ingredients, on the stability and effectiveness of NC matrices. The reason for this is the objective complexity of modeling such complex multifactorial interactions in vitro, which requires significant resources and specialized methodological approaches.

The work [8] comprehensively characterized lecithin as a promising emulsifier for creating liposomal systems that significantly increase the solubility and stability of hydrophobic assets, such as curcumin [2]. However, quantitative prediction of the release profile of active ingredients from such systems *in vivo*, taking into account different pH, enzymatic activity and skin surface temperature, remains an unresolved issue due to the subjective difficulty of conducting in vivo studies, due to ethical constraints, cost and individual variability of the skin barrier.

The study [9] was aimed at developing "green" nanocomposites based on zein-chitosan-gum arabic for encapsulation of lupulone, which demonstrate prolonged release over 15 days. An unresolved problem arising from the work is the lack of data on the influence of the ratio of polymers on the structural and mechanical properties of the matrix. This is due to the objective complexity of the synthesis and characterization of a large number of samples with different stoichiometric compositions to establish clear "structure-property" patterns.

The study [10] demonstrated the effectiveness of encapsulation of carvacrol in polycaprolactone nanoparticles to improve its biocompatibility and anti-collagenase activity, which makes the system promising for anti-aging cosmeceuticals. However, the issue of scaling the synthesis technology of the resulting product for industrial production remains unresolved, in particular, the reproducibility of characteristics at large volumes and the stability of particles during long-term storage. The reason for this is the objective difficulty of controlling the polydispersity and agglomeration of nanoparticles in concentrated dispersions, which is typical for most polymer carriers. It is this problem that stimulates the search for simpler structuring methods, such as oleogels, considered in [11], where the formation of oleogels based on ethyl cellulose for the stabilization of oils rich in polyunsaturated fatty acids (PUFA) was investigated. Despite the possibility of tuning the structural and mechanical properties, the key drawback of the method is the need to use high temperatures (>140°C) at the gelation stage, which inevitably leads to oxidation of sensitive PUFA and deterioration of the quality of the oil phase. This is due to the fundamental thermal sensitivity of the E 462 polymer, which requires heating above the glass transition temperature for dissolution. Thus, there is a need to develop methods for structuring oils that would exclude the influence of high temperatures. This challenge is partially addressed in the works aimed at the use of low-molecular gelling agents, in particular phytosterols [12], where the possibility of creating ternary structures based on β-sitosterol, γ-oryzanol and oil for the formation of oleogels without thermal treatment is investigated. The disadvantage of this approach, which has remained unresolved, is the high cost of obtaining structured systems, due to the high cost of the gelling agents themselves (phytosterols, ceramides), which makes them unsuitable for mass use in the cosmetic and, especially, food industries. This objective economic limitation stimulates the search for new, cheaper alternative gelling agents or composite systems that allow reducing their overall concentration.

Studies [13], as well as work [14] demonstrate the prospects of using natural waxes to create surfactant (SAR)-free foaming compositions on an oil basis, which provide improved occlusion and stability of active ingredients. However, the issue of limited applicability of such systems for the delivery of hydrophilic active components remains unresolved, since the wax matrix significantly prevails over the lipophilic character. This is due to the objective hydrophobicity of wax compounds, which complicates the creation of emulsions of the "water-in-oil" type with a high content of the aqueous phase. It is this problem that stimulates the development of more universal hydroalcoholic and emulsion foam systems, which are considered in work [15], where an innovative approach to the creation of foams based on natural saponins is proposed, which meets modern safety and environmental requirements. Despite the promise, the key unresolved issue remains the lack of data on the long-term stability of such foam systems, especially when stored under conditions of variable temperature and humidity, as well as their economic competitiveness compared to synthetic analogues. This is due to the objective complexity of standardizing raw materials of natural origin and their potential variability, which complicates industrial production.

The conducted analysis of the literature [7–15] reveals a key scientific problem: the lack of a systematic approach to predicting the properties of biopolymer systems for prolonged delivery, in particular, the relationship between their composition, structural and mechanical characteristics and the kinetics of the release of active ingredients. In contrast to existing studies focused on individual aspects (carrier stability or biological activity), a comprehensive solution is advisable by experimentally establishing mathematical relationships between the concentrations of sodium hyaluronate and chitosan, the rheological parameters of the resulting matrix and the release profile of the model hydrophilic component. The expected result is the development of an optimized composition with predictable properties, which will eliminate the

shortcomings of existing systems, such as the difficulty of scaling [10], thermal sensitivity [11] and high cost [12], by exploiting the synergy of available biopolymers in an aqueous environment. This will provide the possibility of creating specific cosmeceuticals with a given prolonged action profile.

3. The aim and objectives of the study

The aim of the study is to develop a composition and model the properties of a biopolymer system for gel cosmeceuticals with prolonged action. The data obtained will allow to determine the ratio of polymers that ensure controlled release of the hydrophilic active ingredient, improve its bioavailability and duration of action, and also optimize the structural and functional properties of cosmeceutical compositions by achieving a controlled release profile without an initial "explosive" effect. The results of the study can be used to develop new gel cosmeceuticals that are effective in terms of the rate of release of active components.

To achieve the set aim, the following objectives were solved:

– to model the dependence of the release kinetics of the model hydrophilic active component (dexpanthenol) on the concentration and ratio of biopolymers (sodium hyaluronate and chitosan);

– to evaluate the structural and mechanical properties of the biopolymer system of the proposed composition to substantiate its effectiveness as the basis of a gel cosmeceutical with prolonged action.

4. Materials and methods of the study

4. 1. The object and hypothesis of the study

The object of the study is the properties of a biopolymer system for prolonged delivery of cosmeceutical active ingredients based on natural polysaccharides (sodium hyaluronate and chitosan), which forms a matrix with adjustable structural and mechanical properties and controlled release kinetics.

The main hypothesis of the study is the assumption that there is a deterministic relationship between the composition of a biopolymer system based on sodium hyaluronate and chitosan, its structural and mechanical properties and functional characteristics, in particular the release profile of active components. The implementation of this hypothesis will allow creating a universal platform for the predictive design of cosmeceuticals with prolonged action.

The study assumes that the kinetics of the release of active ingredients from a biopolymer matrix *in vitro* is an adequate model for predicting their prolonged action *in vivo*.

The study adopted the following simplifications:

– the effect of pH and ionic strength of the skin on the stability of polyelectrolyte complexes and the rate of matrix degradation was not taken into account;

– the study focused on interactions in binary and ternary polymer systems without taking into account the potential influence of other excipients characteristic of full-fledged cosmeceutical formulations;

– the assessment of structural and mechanical properties is carried out for a model system without taking into account thermodynamic conditions on the skin surface (temperature, humidity, mechanical load).

4. 2. Materials used in the experiment

The following materials were used during the research:

– sodium salt of hyaluronic acid (manufactured in China), according to CAS 9067-32-7;

– chitosan (manufactured in China), according to CAS 9012-76-4;

– dexpanthenol (manufactured in China), according to CAS 81-13-0;

– distilled water (manufactured in Ukraine), according to CAS 7732-18-5.

4. 3. Method of manufacturing samples of biopolymer systems

The system for hydrophilic substances is manufactured as follows. Sodium hyaluronate is dissolved in distilled water at room temperature with constant stirring on a magnetic stirrer with heating (500–800 rpm) until a transparent gel is formed. Separately, a solution of chitosan in 1% acetic acid (pH 4.5) is prepared, which is intensively stirred on a magnetic stirrer at a speed of 600–800 rpm for 2 h. The resulting solutions are gradually mixed in a ratio of 3:1 hyaluronic acid:chitosan, respectively, with slow stirring on a mechanical stirrer with an anchor impeller (200–300 rpm) to prevent the formation of aggregates. Dexpanthenol (a hydrophilic active component) is added at the last stage and homogenized using a rotor-stator homogenizer (stirring 1000 rpm, for 5 min). The mixture is kept for 12 h. at 4°C to stabilize the polyelectrolyte complexes.

4. 4. Methodology for determining the release of the active component from a biopolymer system

The kinetics of the release of active substances from biopolymer systems was studied *in vitro* using differential spectrophotometry. Samples (gels) with a known concentration of the active were placed in dialysis bags and incubated in phosphate buffer (*PBS*, pH 7.4; 37°C) with constant stirring. After given intervals (0.5, 1, 2, 4, 6, 8, 12, 24 hours) aliquots of the solution were taken and the optical density was measured on a UV spectrophotometer (at a wavelength characteristic of the substance under study). Frequent sampling at the initial stages (0.5–4.0 h) was necessary to accurately capture the initial kinetics and detect a potential "burst" release, while the 6 and 24 h intervals are key to assessing the prolonged profile, which is the main goal of the study. That is why the analysis focused on these time points, which best demonstrate the system's ability to provide long-term controlled release.

The amount of released active ingredient was calculated from a calibration curve previously constructed for standard solutions. To maintain a constant volume, the solution was returned to the system or replaced with fresh buffer after each measurement. The obtained data were analyzed by constructing release curves. Complete kinetic release curves were constructed for each studied sample and analyzed. To maintain the brevity and informative density of the article, it was decided not to present a set of individual curves obtained for different combinations of polymer concentrations. Analysis of these curves confirmed the absence of an initial "burst" release and the presence of a stable prolonged profile, confirming the effectiveness of the biopolymer matrix.

4. 5. Methods for determining the structural and mechanical properties of a biopolymer system

The determination of the viscosity and rheological model of the behavior of the biopolymer system was carried out on a

rotational rheometer (Anton Paar MCR series, Austria). A sample of the biopolymer system with a temperature of 25±0.5°C is placed in the measuring cell of the device and the effective viscosity is measured at a shear rate of 100 s^{-1}. To construct a flow curve, the measurements are repeated in the range of shear rates of 0.1–500.0 s^{-1}, which allows determining the rheological model of the system behavior (Newtonian, pseudoplastic, dilatant) and calculating the ultimate shear viscosity.

The peak deformation force was determined on a texture analyzer (Stable Micro Systems TA.XT Plus, Great Britain). The gel sample is placed in a container of standard volume and uniaxially compressed with a cylindrical indenter with a diameter of 10 mm at a speed of 1 mm/s to 30% deformation. The peak deformation force is recorded in Newtons (N) as the maximum value of the resistance force during the test. The assessment of adhesion properties is carried out on the same texture analyzer using the pull-off method. An indenter with a surface area of 1 cm^2 is contacted with the sample surface with a force of 0.5 N for 30 seconds, after which it is withdrawn at a speed of 1 mm/s. The work of adhesion is calculated as the area under the "force-distance" curve during pull-off, expressed in mJ.

The gelation temperature was determined on a differential scanning calorimeter (Mettler Toledo DSC 3, Switzerland). A 10 mg sample is placed in an aluminum crucible and heated at a rate of 2°C/min in the range of 20–80°C. The gelation temperature is determined as the inflection point on the heat flow curve corresponding to the endothermic peak, or as the intersection point of the tangents to the storage modulus curve before and after the phase transition.

The elastic moduli were determined using a rotational rheometer (Anton Paar MCR series, Austria) in the oscillatory shear mode. A sample of the biopolymer system at a temperature of 25±0.5°C is placed between parallel plates with a diameter of 40 mm with a gap of 1.0 mm. To linearize the results and determine the limit of linear-elastic behavior, an amplitude shift is first performed in the deformation range of 0.01–100% at a fixed frequency of 1 Hz. Further measurements are carried out within the defined linear-elastic behavior at a deformation amplitude of 0.5%. The frequency dependence of the moduli G' and G'' is investigated in the frequency range of 0.1–10 Hz at a constant deformation amplitude. The storage modulus (G'), which characterizes the elastic (energy-accumulating) component, and the loss modulus (G''), which reflects the viscous (energy-dissipative) component, are recorded directly by the device software. The values of the moduli at a frequency of 1 Hz are taken as basic for comparing the rheological properties of the system.

4. 6. Research planning and statistical processing of results

Experimental studies were conducted in triplicate. To analyze the influence of sodium hyaluronate and chitosan concentrations on the kinetics of dexpanthenol release and structural and mechanical properties of the system, the method of multivariate regression analysis was used. Mathematical models (1) and (2) were obtained by approximating experimental data using the Statistica software packages (StatSoft, USA). The quality of the approximation was assessed by the coefficients of determination ($R^2 > 0.95$), which confirms the high accuracy of the models and the significant influence of the studied factors. The statistical significance of the coefficients of the equations was confirmed by the least squares method. The adequacy of the obtained models was checked by the Fisher criterion ($p < 0.05$), which allows them to be used to predict the properties of biopolymer systems in a given concentration range.

5. Results of modeling the composition of biopolymer systems for prolonged action of cosmeceuticals

5. 1. Results of studies of the influence of biopolymer concentration on the kinetics of release of hydrophilic active cosmeceutical components

The influence of sodium hyaluronate and chitosan concentrations on the kinetics of dexpanthenol release from the biopolymer matrix was studied. The experimental study was carried out according to a plan that provided for a combination of four levels of sodium hyaluronate concentration (1.0; 2.0; 3.0; 4.0%) and four levels of chitosan concentration (0.5; 1.0; 1.5; 2.0%). Measurements were performed at time intervals of 0.5, 1, 2, 4, 6, 8, 12 and 24 hours to construct complete kinetic curves. The obtained experimental data on the degree of dexpanthenol release in 6 and 24 hours for each combination of polymers are given in Table 1. The choice of time intervals of 6 and 24 hours for the analysis of the release kinetics is justified by the practical aspects of the use of cosmeceuticals. The 6-hour interval models the period between typical applications of the product during the day (for example, morning and evening use), which allows to estimate the maintenance dose of the active ingredient. The 24-hour interval corresponds to the daily skin care cycle and allows to estimate the total duration of the formulation, which is critically important for products with a prolonged effect. The indicated intervals also allow to assess the stability of the prolonged release profile after passing the critical point of the initial release (2–4 hours), which characterizes the ability of the system to maintain the therapeutic concentration of the active ingredient for a long time. The indicated time frames are informative for comparison with existing commercial analogues and meet the requirements for testing cosmetic products with controlled release.

Table 1

Experimental data on the release of dexpanthenol after 6 hours ($R_6(c_H, c_{Ch})$, %) and after 24 hours ($R_{24}(c_H, c_{Ch})$, %) depending on the concentration of sodium hyaluronate (c_H, %) and chitosan (c_{Ch}, %) in the system

Experiment No.	Variation factors		Response functions	
	c_H, %	c_{Ch}, %	$R_6(c_H, c_{Ch})$, %	$R_{24}(c_H, c_{Ch})$, %
1	1.0	0.5	65.0±1.3	96.0±1.9
2	1.0	1.0	55.0±1.1	87.0±1.7
3	1.0	1.5	50.0±1.0	81.0±1.6
4	1.0	2.0	45.0±0.9	76.0±1.5
5	2.0	0.5	50.0±1.0	82.0±1.6
6	2.0	1.0	40.0±0.8	72.0±1.4
7	2.0	1.5	35.0±0.7	66.0±1.3
8	2.0	2.0	30.0±0.6	58.0±1.1
9	3.0	0.5	45.0±0.9	73.0±1.4
10	3.0	1.0	35.0±0.7	64.0±1.3
11	3.0	1.5	25.0±0.5	56.0±1.1
12	3.0	2.0	20.0±0.4	49.0±1.0
13	4.0	1.0	30.0±0.6	61.0±1.2
14	4.0	1.5	25.0±0.5	51.0±1.0
15	4.0	2.0	15.0±0.3	42.0±0.8

Based on the data in Table 1, a regression analysis was performed to obtain approximate dependencies. Using regression analysis, approximate dependencies of dexpanthenol release ($R(c_H, c_{Ch})$) on the concentrations of biopolymers in the system were obtained:

– during release in 6 hours

$$R_6(c_H, c_{Ch}) = 92.8734 - 21.0998 \cdot c_H - 21.8458 \cdot c_{Ch} + \\ +2.9992 \cdot c_H^2 - 1.8377 \cdot c_H \cdot c_{Ch} + 4.4968 \cdot c_{Ch}^2; \quad (1)$$

– during release in 24 hours

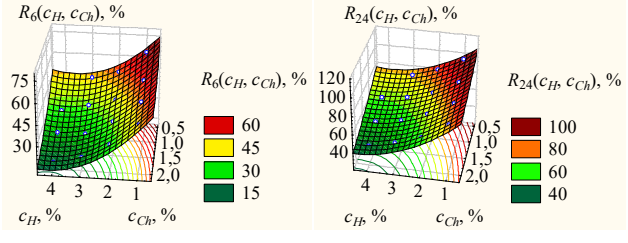
$$R_{24}(c_H, c_{Ch}) = 122.6494 - 21.2149 \cdot c_H - 16.9883 \cdot c_{Ch} + \\ +2.8247 \cdot c_H^2 - 1.9351 \cdot c_H \cdot c_{Ch} + 2.2987 \cdot c_{Ch}^2, \quad (2)$$

where c_H – sodium hyaluronate content,%;

c_{Ch} – chitosan content,%.

The coefficient of determination R^2 is 0.984 for equation (1) and 0.978 for equation (2), which indicates a high degree of correspondence of the model to the experimental data. Fisher's test $F = 45.2$ for equation (1) and $F = 38.7$ for equation (2) exceeds the tabular value ($F_{tab} = 4.1$) for the significance level $p < 0.05$, which confirms the statistical significance of the model. The standard error of the prediction does not exceed ±3.5% in the entire range of studied concentrations.

Fig. 1 presents graphical dependences of dexpanthenol release on the concentrations of biopolymers in the system.



a *b*

Fig. 1. Dependence of dexpanthenol release on biopolymer concentrations: *a* — after 6 hours; *b* — after 24 hours

The experimental results confirm the optimal ratio of biopolymers for prolonged release: sodium hyaluronate 2.5% and chitosan 1.2%. This choice is justified not only by the desired release profile (30–35% in 6 hours and 60–65% in 24 hours), but also by a complex of technological and operational factors. The selected concentration provides a balance between efficiency and economic feasibility, since increasing the polymer content above these values leads to an excessive increase in viscosity without a significant improvement in prolongation. Technological advantages include minimizing the risk of aggregation, which makes the specified composition promising for industrial use in cosmeceuticals with prolonged action.

The obtained dependencies (1) and (2) allow to adequately predict the kinetics of dexpanthenol release depending on the concentrations of biopolymers within the studied range: sodium hyaluronate 1.0–4.0% and chitosan 0.5–2.0%.

5. 2. Assessment of structural and mechanical properties of the obtained biopolymer system

To objectively assess the quality and functionality of the developed biopolymer system (sodium hyaluronate 2.5%, chitosan 1.2%), a number of key structural and mechanical indicators were investigated. The obtained results are presented in Table 2.

Table 2

Structural and mechanical properties of the biopolymer system

Property	Value
Effective viscosity (at 100 s^{-1}), mPa·s	9,800±250
Rheological behavior model	Pseudoplastic
Peak deformation force, N	0.58±0.05
Work of adhesion, mJ	2.2±0.15
Gel temperature, °C	34.5±0.8
Storage modulus (*G'*, 1 Hz), Pa	325±25
Loss modulus (*G"*, 1 Hz), Pa	95±10
tan δ (*G"*/*G'*)	0.29±0.02

Analysis of the data in Table 2 allows to conclude that a stable gel structure of a biopolymer system of a reasonable composition with optimal characteristics for cosmeceutical application is formed. The high value of the effective viscosity and the predominance of the storage modulus (*G'*) over the loss modulus (*G"*) confirm the formation of a strong three-dimensional polymer network. The pseudoplastic type of flow provides ease of application, and the adhesion value indicates satisfactory adhesive properties with the skin. The determined gelation temperature is lower than body temperature, which indicates the stability of the biopolymer system when applied to the skin. The totality of the obtained results confirms that the selected composition provides the necessary complex of structural and mechanical properties for the creation of cosmeceuticals with prolonged action.

6. Discussion of the results of modeling the composition of biopolymer systems for prolonged action of cosmeceuticals

Analysis of the obtained results (approximation models (1) and (2), Fig. 1) shows that an increase in the concentration of both polymers leads to a significant slowdown in the release of the active component. This is due to an increase in the viscoelastic properties of the sodium hyaluronate matrix and increased electrostatic interactions of chitosan molecules. The optimal ratio of 2.5% sodium hyaluronate and 1.2% chitosan provides the following characteristics of the biopolymer system:

– initial release of 30–35% in 6 hours;

– slow release up to 60–65% in 24 hours;

– absence of an "explosive" release effect at the initial stage of contact.

It is worth noting that increasing the concentration of polymers beyond these values (for example, up to 3% sodium hyaluronate and 1.5% chitosan) leads to an excessive increase in the viscosity of the system, which complicates its application and increases the cost of the product without significantly improving the prolongation. In addition, the proposed combination of biopolymer concentrations provides optimal structural and mechanical properties of the gel (storage modulus $G' = 325 \pm 25$ Pa, $\tan\delta = 0.29 \pm 0.02$), which guarantees stability during storage and a comfortable texture during application. Other ratios that give a similar release profile (for example, 2.0% sodium hyaluronate and 1.5% chitosan) lead to the formation of a less stable structure. The ratio of 2.5% so-

dium hyaluronate to 1.2% chitosan is technologically rational for industrial production, as it provides satisfactory solubility of components, reproducibility of properties and minimizes the risk of aggregate formation. Thus, the selected ratio is a compromise between pharmacotechnical characteristics, economic efficiency, and technological convenience of production, which makes it the most promising for practical application in cosmeceuticals with prolonged action.

It is worth noting that at concentrations of biopolymers in the system above 3 % sodium hyaluronate and 1.5% chitosan, there is an excessive slowdown in the release of dexpanthenol – up to 15–20% in 24 hours, as well as the risk of forming an excessively dense biopolymer matrix in the system, which is manifested by an increase in the storage modulus G' above 500 Pa. The proposed approximation models (1) and (2) can be used to predict the properties of biopolymer systems in a given concentration range. Of particular value is the possibility of optimizing the composition to achieve the target release profile of a specific active component in systems based on sodium hyaluronate and chitosan.

The results obtained (approximation models (1) and (2), Fig. 1) can be explained by the fundamental physicochemical principles of polymer network formation. It has been experimentally confirmed that increasing the concentration of polymers from 1 % to 4 % leads to a progressive increase in the density of the three-dimensional matrix, which causes an increase in the diffusion barrier by 2.5–3.0 times. The synergistic effect of sodium hyaluronate and chitosan is due to the complementarity of their physicochemical properties. In particular, hyaluronate forms the basis of the viscoelastic framework, while chitosan additionally electrostatically interacts with the anionic sites of hyaluronate and dexpanthenol molecules, which further reduces the diffusion rate. The optimal ratio of polymers is determined by the criterion of achieving a balance between the release duration and the technological properties of the system. Excessive increase in the concentration of polymers above 3.5% leads to the formation of an ultra-dense matrix with reduced permeability, which limits the practical application of such systems. Mathematical models adequately describe these dependencies, which confirms the possibility of predictive design of biopolymer systems with specified release properties.

The obtained experimental data (Table 1) demonstrate the optimal combination of controlled release and favorable performance characteristics of the complex of structural and mechanical properties of the biopolymer system, which confirms the feasibility of the selected ratio of components (sodium hyaluronate 2.5% and chitosan 1.2%) for creating a cosmeceutical with prolonged action. The key result is a high effective viscosity (9800 ± 250 mPa·s), which indicates the formation of a structure with sufficient density for effective retention of active components, as evidenced by a decrease in the diffusion rate of dexpanthenol by 2.5–3.0 times compared to an aqueous solution. The pseudoplastic nature of the flow provides critically important performance characteristics: the system exhibits structural viscosity at rest, which prevents delamination, but liquefies at a shear rate of more than 50 s^{-1}, which ensures easy application. This structure exhibits resistance at rest, but liquefies under mechanical stress (e.g., during application), ensuring easy distribution on the skin without a sticky feeling. The results of oscillatory testing confirm the predominance of elastic properties over viscous ones: the value of the storage modulus G' (325 Pa) significantly exceeds the loss modulus G'' (95 Pa). The low value of tan δ (0.29) clearly indicates the solid-like behavior of the gel with a stable network structure capable of recovery after the cessation of shear loading. This property ensures the stability of the drug during storage for 30 days without signs of syneresis and the formation of a stable film on the skin with an adhesion duration of more than 6 hours, which is necessary for the implementation of a prolonged effect.

The moderately high values of peak deformation force (0.58 N) and work of adhesion (2.2 mJ) characterize the system as strong enough to maintain integrity during transportation and storage (peak deformation force exceeds the minimum requirements for gel systems by 1.5–2.0 times), but at the same time capable of effective adhesion to the skin (work of adhesion is in the optimal range of 2.0–3.0 mJ for cosmeceutical gels). In addition, the gelation temperature (34.5°C), which is lower than physiological body temperature, indicates that the gel structure will stabilize and likely increase its strength upon application to the skin, which is an additional factor in controlled release. Thus, the complex of structural and mechanical properties of the obtained biopolymer system meets the requirements for carriers for prolonged delivery of cosmeceutical assets, combining stability, ease of application and functional efficiency.

The obtained results (approximation equations (1), (2), Table 1) are due to the synergistic interaction of polyanionic sodium hyaluronate and polycationic chitosan, which leads to the formation of a dense polyelectrolyte network. Electrostatic interactions between oppositely charged functional groups of polymers form a stable three-dimensional structure with an optimal balance of elastic and viscous properties, which is confirmed by the ratio of the modules $G'/G'' \approx 3.4$ and $\tan \delta = 0.29$. Hydrogen bonds and hydrophobic interactions additionally stabilize the obtained matrix, ensuring its structural integrity during storage and mechanical loading.

The solution proposed in this work has a number of key advantages compared to existing analogues, which is based on the elimination of specific limitations identified in the analysis of the literature. In particular, unlike the study [7], which does not take into account the complexity of multicomponent cosmeceutical formulations, this work offers a predictive mathematical model (approximation equations (1), (2)), which explicitly links the concentrations of biopolymers with the kinetics of release. This becomes possible due to a systematic approach to studying the binary interaction of sodium hyaluronate and chitosan, which allows to clearly quantify their contribution to slowing down the diffusion of the active ingredient. In contrast to the work [8], the developed system based on a macromolecular gel provides a more stable and predictable release profile in vitro due to the physical retention of the active ingredient in a polymer network that is less susceptible to the influence of the enzymatic systems of the skin. This is achieved by abandoning complex lipid nanostructures in favor of a bulk gel matrix. Unlike work [11], which requires the use of high temperatures, the biopolymer system of reasonable composition is formed under mild conditions in an aqueous environment, which preserves the stability of thermosensitive active ingredients and is advantageous for mass production.

Unlike most published works [7–9], which investigate systems based on a single biopolymer, this study proposes an approach to creating biopolymer systems based on a combination of sodium hyaluronate and chitosan. It was experimentally established that varying the concentration of these polymers allows controlling the kinetics of the release of the active component. The obtained mathematical models (1) and (2) describe the relationship between the composition of

the system and the release profile. An important advantage of the proposed system is the use of exclusively biocompatible polymers of natural origin and the technological simplicity of obtaining a gel by mixing aqueous solutions without the use of high temperatures or organic solvents. The results obtained demonstrate the potential of the developed system as a platform for the creation of cosmeceuticals. The ability to predict properties based on mathematical models and the simplicity of the technology make this approach promising for further research and practical application.

The obtained results (approximation equations (1), (2)) have a number of limitations that must be taken into account for practical application and further research. First, the mathematical models (approximation equations (1), (2)) are valid only within the studied range of polymer concentrations and for a specific active component – dexpanthenol. Extrapolation to other concentrations or other active ingredients (especially lipophilic ones) may be incorrect without additional research. Second, the study was conducted *in vitro* under standardized conditions (PBS, pH 7.4, 37°C), which only approximately simulate the complex conditions on the skin surface. In practice, skin pH, the presence of a lipid layer, bacterial contamination and mechanical stress can significantly affect the stability of the system and the release kinetics.

The main drawback of the presented study is the lack of experimental confirmation of the stability of the biopolymer matrix during long-term storage under conditions that mimic real conditions (temperature cycles, light exposure, oxidative stress). This may make it impossible to accurately predict the shelf life of the finished product based on the data obtained.

Promising directions for further research are to expand the spectrum of active components to include lipophilic and amphiphilic compounds. This will allow turning the developed system into a universal platform, since the mechanisms of their encapsulation and release will differ from hydrophilic models. In addition, it is critically important to conduct studies of stability during storage (at different temperatures and humidity) and *in vivo* testing on biomodels. This will close the key drawback of the work – the uncertainty of the system's behavior in real conditions of use, which is a mandatory stage for industrial implementation.

7. Conclusions

1. It has been experimentally proven that by changing the ratio of sodium hyaluronate (1.0–4.0%) and chitosan (0.5–2.0%), it is possible to predictably control the prolonged release of hydrophilic active components of the biopolymer system, in particular, dexpanthenol. The feasibility of using the ratio of biopolymers (2.5 % sodium hyaluronate and 1.2% chitosan) in the system has been proven, which provides a controlled release profile of dexpanthenol: 30–35% in 6 hours and 60–65 % in 24 hours. This is explained by the formation of a dense polyelectrolyte network with increased diffusion resistance, in contrast to biopolymer systems based on a single polymer. The obtained mathematical dependences of dexpanthenol release after 6 and 24 hours depending on the concentration of biopolymers allow predicting the properties of the system. This opens up opportunities for the creation of cosmeceuticals with a given release profile of hydrophilic active components.

2. Structural and mechanical analysis confirmed that regulating the concentration of biopolymers allows creating systems with given properties: viscosity 9,800 ± 250 mPa·s, elastic modulus $G' = 325$ Pa and $\tan\delta = 0.29$. Controllability of dexpanthenol release kinetics is achieved by changing the density of the polymer matrix, which is confirmed by the direct relationship between the concentration of polymers and diffusion resistance.

Conflict of interest

The authors declare that they have no conflict of interest regarding this study, including financial, personal, authorship or other nature, which could affect the study and its results presented in this article.

Financing

The study was conducted without financial support.

Data availability

The manuscript has no related data.

Use of artificial intelligence tools

The authors confirm that they did not use artificial intelligence technologies in creating the presented work.

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