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Usage of Mathematical Modeling and Optimization in Development of Hydrogel Medical Dressings Production

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Abstract: In connection with the significant complication of research objects of technological systems and the considerable increase in expenses for carrying out experimental research, improving the mathematical modeling methods of these systems is a current problem. By using the means of mathematical modeling and optimization, the calculation of the main technological parameters of the formation method of film hydrogel products based on silver-filled copolymers of 2-hydroxyethylmethacrylate with polyvinylpyrrolidone was performed. The technological parameters of the polymerization processes, chemical reduction of silver ions, and centrifugal formation of the film cloth were substantiated. These are the components of the technological process, which occurs in one stage in the form of a centrifugal unit. By using the obtained results, silver-filled films were obtained, which are characterized by unique properties and can be used in the treatment of trophic ulcers of lower limbs.

Keywords: mathematical modeling; technological parameters; hydrogel medical dressings; silver-containing hydrogels; centrifugal formation; trophic ulcers

1. Introduction

Modern methods of mathematical modeling, synthesis, and optimization of technological parameters of production systems anticipate, in particular, a set of different methods of a qualitative [1] and quantitative [2,3] nature.

Currently, significant success has been achieved in the development of new methods and the optimization of synthesis conditions and properties of composite polymers [4–9]. This is a prerequisite for creation materials with unique properties for different fields of application [10–13]. Especially as it concerns the composite polymer hydrogels [14–17], among which the most popular in science and practice are hydrogels filled with (nano) metal particles [18–20].

The uniqueness of such materials is the combination of the properties of a polymer matrix and metal filler. A polymer matrix is characterized by the ability to absorb low molecular substances, including medicines; to swell in solvents; and to retain a significant amount of water while being in a highly elastic state. Depending on the nature of metal, composite hydrogel can acquire electro-conductive, magnetic, and anti-bacterial properties, which significantly expand the fields of its usage [20–23]. Composite metal hydrogels with antibacterial and antifungal properties are ideal materials in the medical field for the creation of dressings to treat wounds, burns, and ulcers of various kinds, including venous ulcers of lower limbs [24–26].

The modern practice of the treatment of venous ulcers of lower limbs implies a combination of various methods for conservative treatment and surgical interventions.

Most trophic venous ulcers are characterized by a high degree of bacterial colonization with high probability of developing wound infection. According to many researchers, an important condition necessary to prevent recurrence of venous ulcers is the application of adequate systemic and local therapy with antibiotics and antiseptic preparations in combination with lower limb compression [27].

One of the promising methods for the treatment of venous ulcers is considered to be covering them (in order to prevent the development of wound infection) with sorption hydrogel film materials with antibacterial properties and the combination of therapy using medical preparations [28].

We established the prospects of using silver-filled film hydrogel products based on co-polymers of polyvinylpyrrolidone (PVP) with 2-hydroxyethylmethacrylat (HEMA) during the treatment of venous ulcers [25].

High elasticity, strength, sorption capacity, and bactericidal and antifungal properties of the obtained materials based on co-polymers of polyvinylpyrrolidone with 2-hydroxyethylmethacrylat (PVP-gr-pHEMA) make them effective to use as hydrogel dressings for medical purposes. A unique porous structure [17], combined with the existence of hydrophilic functional groups, ensures swelling of a polymer matrix in water and high permeability for dissolved low-molecular substances. In turn, this determines the suitability of the obtained hydrogel dressings for the preparation treatment by introducing medicines through the material by the transdermal method [25].

At the same time, due to the ability to absorb and retain moisture, elasticity, and stability of the form in an aqueous medium, such materials are compatible with a variety of biological systems.

2. The Analysis of Previous Research and Problem Statement

A new method was proposed for obtaining metal-filled hydrogels based on copolymers PVP with HEMA. The method consists of implementation of polymerization with the simultaneous reduction of metal ions by using exoeffect of polymerization [29].

The method is especially attractive, both from the practical and the scientific point of view, because the particles of metal are formed at the same time as the formation of a polymer matrix. This approach makes it possible to achieve a better, more uniform distribution of the filler and to obtain the material with the isotropic properties.

High reactivity and the possibility to regulate, within wide limits, the time of PVP/HEMA compositions in a liquid state is a prerequisite of their recycling in film products by the method of centrifugal formation [25]. The technological characteristics [30], cost effectiveness of the centrifugal formation, and low quality of the films obtained by other existing methods contribute to this (casting and filling into glass or polymer forms). The samples of hydrogel film materials obtained by centrifugal formation are characterized by various thicknesses, which do not exceed 1%, high-quality surfaces, and a complex of physical and mechanical properties.

Centrifugal molding is one method of producing plastic products in the bodies of revolution by the filling of the material in a viscous-flow state into the form that rotates in the same plane. Material is evenly distributed centrifugally on the inner surface of the form in a continuous layer while receiving curing-required configuration [31].

The proposed technology is easy to implement in production and does not require a sophisticated apparatus design. One of its advantages is the minimum amount of waste, so there is no need of recycling or recuperation of solutions of an oxidant and a reducing agent. Such technology is a priority and needs development, taking into consideration the prospects of the obtained composites.

However, as for any technology, one of the main factors in obtaining high-quality hydrogel film products is the optimal technological parameters of the technological manufacturing process. At the same time, the establishment of optimal technological parameters requires implementing a large number of experimental studies and the use of mathematical modeling and forecasting methods.

Developed methods of optimization provide the use of mathematical apparatus not only in analyzing research results but also during their forecast, which provides an opportunity to significantly decrease materials and time consumption in the experiment [32,33].

Therefore, the aim of this study was to establish the technological parameters of obtaining hydrogel films for medical purposes by using the means of mathematical modeling. The films were obtained by the centrifugal method with the simultaneous formation of a polymer matrix and obtainment of metal filler particles.

3. Object of Investigation

Lightly crosslinked polymers obtained by radical polymerization of HEMA in the presence of PVP were selected as a matrix for filling. High-purity polyvinylpyrrolidone (AppliChem GmbH, Darmstadt, Germany) with MM 12,000 and 2-hydroxyethylmethacrylat (Sigma Chemical Co., Burlington, MA, USA) were used for co-polymerization. The initiator of the radical type benzoyl peroxide (BP) was selected as the polymerization initiator. This choice was determined by its wide use for the synthesis of pHEMA-gr-PVP co-polymers [20]. HEMA was distilled in a vacuum (residual pressure was 130 N/m^2, T_{boil} = 351 K); PVP was dried at 338 K in a vacuum for 2–3 h; and BP was re-crystallized from ethanol. Polymerization was carried out with the initiator of a radical type of BP (T_0 = 50 °C). Given that the silver-filled hydrogel is meant for medical purposes, silver deposition was carried out from argentum nitrate (of chemically pure brand) in the aqueous-ethanol solution [34].

4. Calculation Results of Technological Parameters of the Obtaining Process of Hydrogel Films

Technological parameters are a set of conditions that ensure the obtainment of a quality product. Each parameter has a greater or lesser effect, but this influence is interrelated, so in the technology, the optimal set of parameters should be established. Deviation of one of them can lead to changes in product quality (both in terms of physico-mechanical and performance characteristics, and in terms of appearance) and process productivity.

For the formation of hydrogel films, a centrifugal unit was designed with the main element of centrifugal mold (Figure 1).
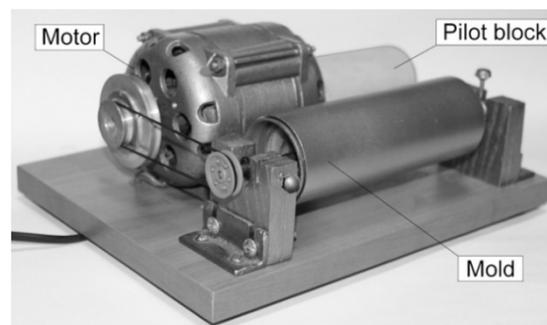

Figure 1. Installation for centrifugal molding of composite films from polymer hydrogels.

The developed obtainment method of silver-filled hydrogel films by polymerization with simultaneous reduction of Ag$^+$ ions anticipates the production of films in one stage on one piece of equipment. The method is peculiar in that, during the time it takes to form the final product—the time between loading the reaction composition and removing the final product in the centrifugal form—three processes that differ in nature occur at the same time: the chemical process (synthesis of a polymer matrix due to copolymerization of PVP with HEMA); Ag(0) formation by reduction of Ag$^+$ ions; and the physical process (acquisition of a film cloth by centrifugal molding). Each process must be carried out under certain conditions, which form a set of technological parameters of the technological obtainment process of hydrogel films (Figure 2).
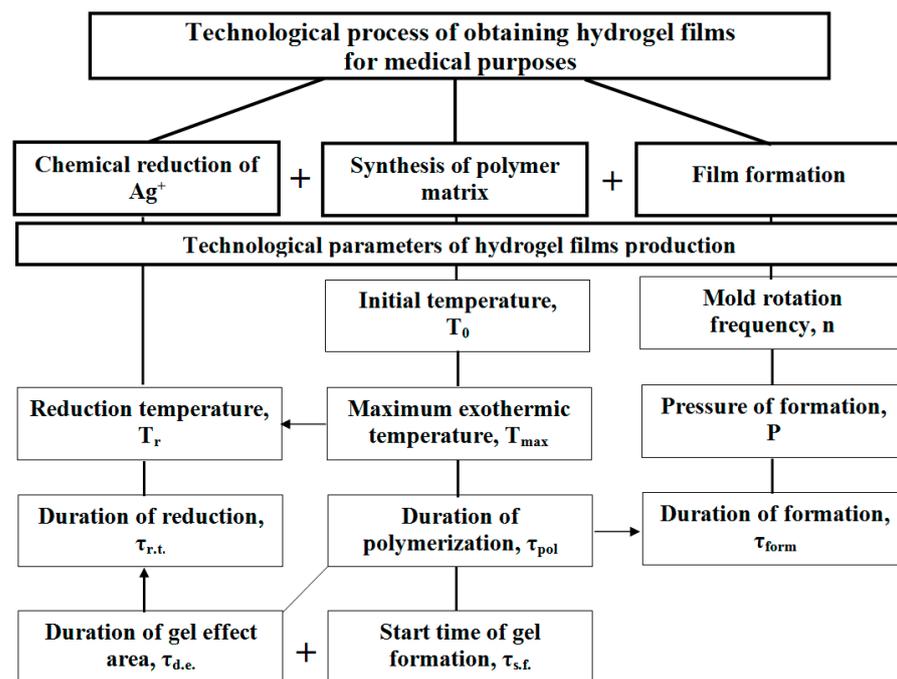
Figure 2. Technological parameters of hydrogel films production.

As kinetic investigations of the reduction of the speed of ions have shown, the main parameters of the process are the duration of the induction period of reduction ($\tau_{i.r.}$), the duration of reduction ($\tau_{r.t.}$), and the reduction temperature (T_r) [28]. A start time of gel formation ($\tau_{s.f.}$), a duration of gel-effect area ($\tau_{d.e.}$), and the maximum exothermic temperature (T_{max}) [28,29] were used for a characteristic of the copolymerization process PVP with HEMA [26,27]. The parameters $\tau_{s.f.}$, $\tau_{d.e.}$, and T_{max} were taken on the basis of thermometric polymerization research [28,29].

In our case, it is important that the duration of the Ag^+ reduction process approaches the duration of the gel-effect area of the polymerization reaction with a minimum start time of gel formation and a maximum T_{max}. The duration of molding is determined by the duration of polymerization, which depends, in our case, mainly on the formulation of the original reaction composition and is established by the experimental method.

This method requires the selection of compositions that polymerize with the maximum heat liberation, the minimum induction period, and the maximum gel-effect area according to the technological features of obtaining metal-filled hydrogels.

In this experiment, the optimal content of the initial composition components that achieve the necessary parameters of the exothermic process could not be found. Therefore, for investigation of the simultaneous influence of initial composition components on the exothermic process parameters, we optimized the experiment by means of simplex-lattice planning (Scheffe's method) in order to reduce the cost of the experiment [35]. The result of the research is a multifactor mathematical model in the form of a polynomial of a given degree. The necessary condition in the simplex method is to provide at each experimental point a condition fulfillment $\Sigma X_i = 1$, where $X_i \geq 0$—the concentration of the *i*-th component in the composition. During the research of the mixtures properties, which depend only on three components, the factor space is an equilateral triangle, and for the system, the ratio is executed: $X_1 + X_2 + X_3 = 1$ [35]. The vertices of the triangle correspond to the pure substances, and the sides correspond to the double systems. In our case, the whole concentration triangle was not investigated, only its local part, which is a simplex with vertices A_1 (72% HEMA; 8% PVP; 20% H_2O); A_2 (56% HEMA; 24% PVP; 20% H_2O); and A_3 (56% HEMA; 8% PVP; 36% H_2O) (Figure 3a), where X_1 denotes HEMA, wt.%; X_2 is PVP, wt.%; and X_3 is H_2O, wt.%.
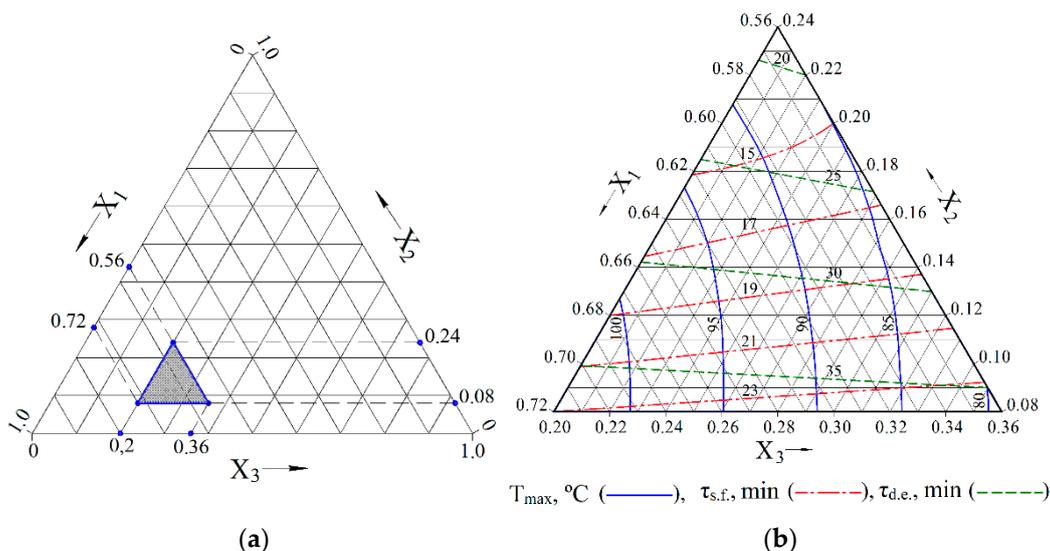
Figure 3. Triangles for the research of "composition-technological parameters" diagrams: (**a**) local area of research; (**b**) equal values lines of parameters of the exothermic process.

The optimization for exothermic parameters—the start time of the gel formation ($\tau_{s.f.}$, min), gel-effect area ($\tau_{d.e.}$, min), and the maximum exothermic temperature (T_{max}, °C)—was performed. In Table 1, conditions and results of experiments in the form of pseudo-components and on a natural scale are presented. The average results, y_1^e (T_{max}, °C), y_2^e ($\tau_{s.f.}$, min), and y_3^e ($\tau_{d.e.}$, min), were obtained by two parallel experiments.

Table 1. Conditions and results of experiments.

№	Natural Variables			y_1^e	y_2^e	y_3^e
	X_1	X_2	X_3			
1	0.72	0.08	0.20	103.5	23.00	37.00
2	0.56	0.24	0.20	84.40	13.00	18.20
3	0.56	0.08	0.36	79.00	24.20	36.50
4	0.64	0.16	0.20	96.80	16.00	28.00
5	0.64	0.08	0.28	92.00	23.70	36.90
6	0.56	0.16	0.28	84.00	17.40	26.40

Using the matrix of planning and conditions and results of the experiment (Table 1), the coefficients of the polynomial were calculated, and the regression equations were derived [36]:

$$y_1 = 123.44 + 63.75X_2 - 71.25X_3 - 203.13X_2X_3 - 445.31X_2^2 - 117.19X_3^2; \tag{1}$$

$$y_2 = 33.13 - 184.38X_2 + 7.5X_3 + 109.38X_2X_3 + 312.5X_2^2 - 15.63X_3^2; \tag{2}$$

$$y_3 = 40.39 - 50.63X_2 + 28.75X_3 - 234.38X_2X_3 - 62.5X_2^2 - 23.44X_3^2. \tag{3}$$

The obtained equations make it possible to predict the parameter change character of the exothermic copolymerization process for PVP with HEMA: the start time of gel formation (y_1), the gel-effect area (y_2), and the maximum exothermic temperature (y_3) for any composition of the initial composition. According to the obtained regression equations, the isolines of the exothermic parameter change were plotted depending on each component content of the original composition (Figure 3b). The obtained lines of equal parameter values provide a quick search for a totality of values of the components concentrations in the reaction composition, which makes it possible to obtain the optimal technological conditions that are necessary for the metal deposition during the polymerization.

The main technological parameters of the centrifugal molding are rotational frequency of mold, molding pressure, and centrifugal force. The frequency of mold rotation depends on the quality, physical, and mechanical properties of the film. Rotational frequency affects the magnitude of the centrifugal force and, accordingly, the molding pressure and density of the film. In this work, the functional connection was established, which makes it possible to determine the parameters and the relationship between them.

To calculate the centrifugal force F_c acting in a cylindrical mold on PVP/HEMA composition, we used the following equation (on the basis of Newton's second law):

$$F_c = \frac{mv^2}{R}, \tag{4}$$

where m = mass of the load to the form composition, down kg; v = linear velocity, m/s; R = radius of the inner mold surface (outer radius of the product), m (Figure 4).
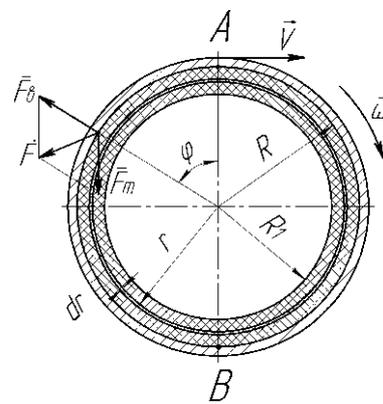

Figure 4. Cross-section of a cylindrical mold with a composition and location of existing forces during centrifugal molding.

Linear velocity is determined through the angular velocity of the following equation:

$$v = \omega \times R, \tag{5}$$

where ω = angular velocity, s^{-1}.

The angular velocity is related to the frequency of revolutions n (s^{-1}):

$$\omega = 2 \times \pi \times n, \tag{6}$$

Using Equations (4)–(6) we obtained

$$F_c = 4 \times \pi^2 \times m \times n^2 \times R \text{ or } F_c = 2 \times \pi^2 \times m \times n^2 \times D, \tag{7}$$

From Equation (7), we see that the centrifugal force acting on the composition in a cylindrical mold depends on the frequency and the diameter of the designed cavity. As the centrifugal force is the main factor that affects the quality of products by centrifugal molding, the nature of filling out a mold, namely the distribution of material on the surface of the designed form, depends on the frequency of rotation, i.e., the angular velocity. Thus, in low rate, the composition will rise to some height in a turbulent wall layer in the direction of rotation and flow down to the bottom wall (Figure 5a).

With increasing rotational frequency of mold compositions, lifting height increases. For a certain number of revolutions, composition reaches a certain point and then falls down (Figure 5b). Film formation in this case is also impossible. With the further increase in angular velocity, forms of the centrifugal force acting on the material take the value at which the composition begins to rotate with the mold without detaching from its inner surface (Figure 5c). Rotational frequency of the mold in this case is called critical n_{cr}.

In order to form high-quality surface films, the working rotational frequency of mold n_w must be greater than critical:

$$n_w > n_{cr}, \tag{8}$$

Figure 4 demonstrates the forces acting on the composition during the molding of the product. Vector centrifugal force is directed along the radius from the center; the vector of gravity acting on the material is directed down perpendicular to the axis of rotation of the shape. Using the cosine theorem, we obtain

$$F^2 = \left(mR\omega^2\right)^2 + (mg)^2 - 2m\omega^2 R \times mg \times \cos\phi, \tag{9}$$

where $\phi = 0°$ (point A) $\cos\phi = 1$, then

$$F_c = \sqrt{(mR\omega^2 - mg)^2} = mR\omega^2 - mg, \tag{10}$$

where $\phi = 180°$ (point B) $\cos\phi = -1$, then

$$F_c = \sqrt{(mR\omega^2 + mg)^2} = mR\omega^2 + mg, \tag{11}$$

Thus, when the polymer–monomer composition is at point B, it acts with more force than at point A. This distributes the material uniformly over the inner surface of a horizontal form. From Equation (10), we see that the composition moves along the mold surface at a certain value (critical) centrifugal force, which is greater than the force of gravity:

$$F_c > F_g, \text{ i.e., } mR\omega_{c,r}^2 > mg, \tag{12}$$

Consider that $\omega = 2 \times \pi \times n$:

$$mR4\pi^2 n_{cr}^2 > mg \text{ and } n_{cr} > \sqrt{\frac{g}{4\pi^2 R}}, \tag{13}$$

Of course, the quality of the film surface primarily depends on the pressing force of forming film to the surface. The pressure that causes the centrifugal force on the composition layer is equal to:

$$P = \frac{F_c}{S} = \frac{F_c}{2\pi Rl}, \tag{14}$$

where $S = 2\pi Rl$ = surface area of the composition layer, m²; l = work length of designed mold, m.

Mass of the elementary layer dm can be calculated as

$$dm = \rho \times 2\pi \times r \times l \times dr, \tag{15}$$

where ρ = density of the composition, kg/m³; r = radius of the elementary layer of the composition, m; dr = the thickness of the elementary layer of composition, m.

Then, the magnitude of the centrifugal force acting on an elementary layer of radius r and thickness dr (Figure 4) can be represented as

$$dF_c = \frac{\rho \times 2\pi \times r \times l \times dr \times \omega^2 \times r^2}{r} = 2\pi \times l \times \rho \times \omega^2 \times r^2 \times dr, \tag{16}$$

Integrating Equation (16) from R_1 to R, we obtain the equation for determining the centrifugal force acting on the composition:

$$F_c = 2\pi l\rho\omega^2 \int_{R_1}^{R} r^2 dr = \frac{2\pi l\rho\omega^2 \left(R^3 - R_1^3\right)}{3}, \tag{17}$$

where R = outer diameter of the film, m; R_1 = inner diameter of the film, m.

From Equation (14) using (17), we determine the pressure acting on the outer surface of the film:

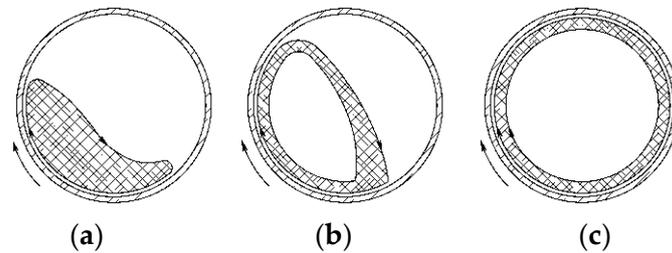$$P = \frac{\rho\omega^2(R^3 - R_1^3)}{3R} \text{ or } P = \frac{4\rho\pi^2 n^2(R^3 - R_1^3)}{3R}, \tag{18}$$

(a)　　　　　　　**(b)**　　　　　　　**(c)**

Figure 5. Routing of the composition in a cylindrical mold at different rotational frequency: $n_w \ll n_{cr}$ (**a**), $n_w \leq n_{cr}$ (**b**), $n_w > n_{cr}$ (**c**).

5. Implementation of the Obtained Results

By using these results, experimental samples of composite hydrogel films were obtained (Figure 6). Hydrogel film material samples obtained by centrifugal molding have attracted attention because of the film's high surface quality and its complex of physico-mechanical properties, and because the isopach does not exceed 1%.

Figure 6. Experimental sample of hydrogel film.

The medical–biological studies of the resulting film products were carried out under laboratory conditions at the Department of Microbiology of Danylo Halytskyi Lviv National Medical University. A comparative analysis of the results of medical–biological tests of the obtained materials and non-filled hydrogel films regarding the used micro-organisms revealed that non-filled films do not show any bactericidal or antifungal properties. The film products that contain silver particles block the growth of bacteria and fungi (Table 2) [25].

Table 2. Bactericidal and antifungal activity of silver-filled hydrogel films, obtained based on copolymers of polyvinylpyrroli-done (PVP) with 2-hydroxyethylmethacrylat (HEMA).

Duration of Storage of Hydrogel Films	Magnitude of the Zone of Inhibition of Microorganism Growth, mm				
	S. aureus	*S. epidermidis*	*Str. viridans*	*E. coli*	*C. albicans*
1 month	12, 10, 9	12, 8, 9	11, 11, 9	3, 3, 0	11, 11, 7
18 months	5, 4, 8	5, 4, 5	11, 8, 8	–	0, 4, 2

Based on the obtained silver-filled films, we developed hydrogel medical dressings, and the clinical testing of which was successfully carried out at the surgical department

of the Clinical Hospital of Lviv Railway in the treatment of venous ulcers of lower limbs. It was established that the use of the silver-filled hydrogel films improves treatment results; accelerates cleaning, granulation, and healing of trophic ulcers; and, as a result, reduces the duration of patients staying at hospital. Due to their unique properties, the developed materials can be also used for the treatment of burns and post-operative wounds.

6. Conclusions

The basic technological parameters of formation processes of a film hydrogel cloth by a centrifugal method were calculated. The technological parameters of the polymerization processes, chemical reduction of silver ions, and the centrifugal method were established. They compile the technological parameters of the technological obtainment process of hydrogel dressings for medical purposes.

Using the Scheffe's simplex-lattice planning method, the experiment was optimized to forecast the polymerization parameters of PVP/HEMA compositions, which define the technological regime of metals' chemical precipitation.

The results of the clinical studies show sufficient clinical effectiveness of using the developed hydrogel dressings for medical purposes based on hydrogels containing silver particles. Such materials in combination with the integrated therapy help to increase the speed and effectiveness of treatment of trophic venous ulcers of lower limbs.

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