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Software for Experiment Planning and Modelling in Research of Nanofilled Three-Component Systems

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Software for automated interactive planning of experimental investigations of three-component heterogeneous systems, according to the given restrictions on the content of components, is developed. The program can be used for all possible combinations of ingredients in the composition. For mixtures, in which the content of one or two of the components is less than the others by several orders of magnitude, the created software has developed algorithm for unevenly increasing the subarea of planning with the mandatory preservation of mathematical co-ordinates. The program allows us to design an experimental plan, using one of three types of models of dependence of the output parameters on the content of components, namely, quadratic, incomplete cubic and cubic ones. Using a mathematical model based on an incomplete third-order polynomial, the optimal content of alumina nanoparticles and carbon nanotubes (CNT) for the modification of polypropylene monothreads is calculated. As established, the introduction of 1.1 wt.% Al_2O_3 and 0.5 wt.% CNT into the composition provides the threads with increased strength and elasticity, as well as high antimicrobial action against test-strains of microorganisms and fungi.

Розроблено програмне забезпечення (ПЗ) для автоматизованого інтерактивного планування експериментальних досліджень трикомпонентних гетерогенних систем за заданими обмеженнями на вміст компонентів. Програма може бути використана для всіх можливих комбінацій інґредієнтів у композиції. Для сумішей, в яких вміст одного або двох із компонентів є меншим за інші на декілька порядків, у створеному ПЗ розроблено алґоритм для нерівномірного збільшення підобласти планування з обов'язковим збереженням відповідности математичних координат. Програма уможливлює будувати план експерименту, використовуючи один із трьох типів моделів залежности вихідних параметрів від вмісту компонентів — квадратичної, неповної кубічної та кубічної. За допомогою математичного моделю, створеного на основі полінома неповного третього порядку, розраховано оптимальний вміст наночастинок оксиду Алюмінію та вуглецевих нанотрубок (ВНТ) для модифікування поліпропіленових монониток. Встановлено, що введення в композицію 1,1 мас.% Al_2O_3 та 0,5 мас.% ВНТ забезпечує ниткам підвищені міцність та еластичність, а також високу антимікробну дію до тест-штамів мікроорганізмів і грибів.

Key words: plan of experiment, software, mathematical model, nanoadditive, composition.

Ключові слова: план експерименту, програмне забезпечення, математична модель, нанодобавка, композиція.

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1. INTRODUCTION

At the beginning of the third millennium, polymer composite materials became important in various fields of human activity, revolutionizing the technique, everyday life, and lifestyle. Areas of their application are expanded from household goods (fabrics, textiles, knitwear, packaging, biomedical sparrows) to high-tech products (aerospace and military equipment). This is due to a number of their advantages over other materials, namely, thermal and corrosion resistance, lightweight combined with high mechanical properties, low cost. Providing polymer products with a set of desired characteristics is achieved through a wide range of existing modification methods: mixing, copolymerization, introduction of additives (plasticizers, stabilizers, compatibilizers, fillers, *etc.*).

Qualitatively new materials with improved adjustable properties are obtained by a combination of polymer matrices with nanofillers of different chemical nature, configuration and size. The main factors, which determine the characteristics of nanocomposites, are both the chemical nature of the polymer and additives and the ratio of ingredients in the mixture. Thus, the introduction of highly dispersed electrically-conductive fillers in incompatible mixtures of polymers can reduce the concentration of the additive required to reduce the percolation threshold due to the selective localization of nanoparticles (NP) in the transition layer [1]. When obtaining microporous polymeric materials by the template method (selective removal of one of the components from nanofilled composites), materials with adjustable porous structure are obtained. The values of porosity and average pore diameter in polyethylene membranes were regulated by changing the number of particles of organomodified montmorillonite in the composite film [2]. When adding of organoclay to the mixture polyamide/polylactide (PA/PLA), there is a change in the type of PA structure in the matrix, namely, the transition from drip-matrix to intertwined morphology. This led to an increase in heat resistance and plasticity of the composite material without adversely affecting its rigidity and strength [3]. By varying the content of nanoparticles of alumina and titanium dioxide in a mixture of polypropylene/copolyamide (PP/CPA) and polyethylene terephthalate (PETF)/PP, respectively, it is improved the performance of composite threads, namely, increasing their strength and shape resistance by reducing the diameters of microfibrils PP and PETF formed *in situ* [4, 5]. The characteristics of complex threads from microfibrils obtained by extraction of the matrix polymer from composite filaments also depend on the content of the nanoadditive. The values of tensile strength, modulus of elasticity, specific surface area, hygroscopicity have maximum values at a certain concentration of alumina [4], silica [6] or mixed oxide (TiO_2/SiO_2) [7]. With a higher content of additives, the properties of composite and complex threads are deteriorated that is the result of saturation of the interphase zone with nanoparticles. When studying the effect of clay on the dimensional characteristics of natural rubber particles in the PP matrix, the effect of reducing the surface activity of the filler, when reaching its appropriate concentration, was also observed [8].

Synthetic filaments filled with NP of precious metals and metal oxides have a wide range of antimicrobial action, have antiallergic, sorption and antistatic properties, protect from UV radiation, *etc.* [9, 10]. The combined use of two substances in the nanostate is more effective. Thus, with the simultaneous introduction of nanoparticles of silver and copper, the bactericidal characteristics of the threads are higher, compared with individual additives, due to the mutual synergistic enhancement of their action [10]. Electroforming polyvinyl alcohol nanofibers, filled with NP of bicomponent Ag/TiO_2 additive, have a high specific surface area (200 m^2/g). The presence of silver provides them with antimicrobial activity, and TiO_2 provides photocatalytic activity [11]. The introduction of silver/silica nanoadditives into the structure of polypropylene monothreads gives them biological activity and improves mechanical and manipulative properties [12].

Studies of mixtures' compositions are associated with the need to perform a large number of multifactorial experiments (even without parallel experiments); so, it is advisable to use mathematical planning methods. At the same time, various mathematical functions are used to describe the properties of the system, which relate the output parameters to the variables during the experiments: $y = \phi(x_1, x_2, x_3)$. The response surfaces for three-component compositions are adequately described by polynomials of high degrees, but they have a large number of coefficients (in a polynomial of degree n for q components, C_{q+n}^{n} coefficients are accrued). To accelerate and increase the reliability of research results, it will help the development of software to create a working plan of experiments in order to obtain the highest-quality mathematical model with the least possible number of experiments. An important factor is the optimization of the content of ingredients in the composition, as it is necessary that the desired properties of the products be achieved with a minimum content of modifiers. This also solves the problem of reducing the toxic effects of nanoadditives on living organisms and the environment and reduces the cost of production.

The purpose of the work is to develop software for creating a working plan of experiments and a mathematical model, as well as to calculate the optimal composition of a three-component mixture to obtain polypropylene threads with high antimicrobial and mechanical properties.

2. MATERIALS AND METHODS

The software for development of a working plan of the experiment for study of three-component compositions is created in Builder environment in C++ [13–15]. For planning, it was used the simplex-lattice method due to its simplicity and efficiency in the study of multicomponent compositions [16]. The variables x_i ($i = 1, 2, \ldots, q$) of such systems are the proportions (relative to the content) of the i-th components of the mixture and satisfy the condition:

$$\sum_{1 \le i \le q} x_i = 1 \ \ (x_i \ge 0)\,. \tag{1}$$

The geometrical place of points satisfying condition (1) is a $(q-1)$-dimensional simplex, which, for a three-component system, has the form of an equilateral triangle. Each point of such a simplex corresponds to a mixture of a certain composition and, conversely, any combination of relative contents of the q components corresponds to a certain point of the simplex.

The problem of constructing a mathematical model 'composition–property' is solved by representation the desired function in the form of the polynomial of degree n in canonical form. To estimate the coefficients of the polynomial, plans have been proposed, which provide a uniform scatter of experimental points over the $(q-1)$-dimensional simplex [16]. The points of such plans are nodes of $\{q, n\}$-simplex lattices, in which, for each factor (component), $(n+1)$ evenly spaced levels in the range from 0 to 1 ($x_i = 0, n^{-1}, 2n^{-1}, \ldots, 1$) are used, and their different combinations are taken. The set of points $(x_{1u}, x_{2u}, \ldots, x_{qu})$, where $u = 1, 2, \ldots, N$, forms a saturated simplex-lattice plan.

Experimental studies were performed using isotactic polypropylene brand Sabic 575P with a melt-flow rate of 11 g/10 min and a melting point of 169°C. Nanoparticles of pyrogenic alumina with a specific surface area of 109 m^2/g and the three-layer carbon nanotubes (CNT) with an outer diameter of 10–20 nm and a specific surface area of 340 m^2/g were selected as nanoadditives. The ingredients of the composition were mixed on a worm-disc extruder. Nanofilled polypropylene monothreads were formed on a laboratory stand through a die with a diameter of 780 μm at a temperature of 190°C, with a drawing degree of 1000%. Thermoorientation drawing was carried out at a temperature of 150°C with a multiplicity of 9.5. The tensile strength of the threads was determined using a rupture machine brand KT 7010 AZ. Studies of bactericidal properties of the threads were performed on WHO-recommended test-strains of microorganisms and fungi, according to standard methods [17]. The antimicrobial activity of the modified PP threads was determined by the diameter of the zone of growth retardation of microorganisms (*D*, in millimetres).

To build the plan of experiment, the content of individual ingredients of the three-component system was subject to bilateral restrictions $0 \le a_i \le x_i \le b_i \le 1,\ i = \overline{1, q}$, where a_i and b_i are the upper and lower limits of the restrictions for each of the components, which should not be equal to each other. For the studied composition, the input variables were x_1, x_2, x_3—relative concentrations of PP, nanoparticles of Al_2O_3 and CNT, respectively, which were subject to the following restrictions:

$$0.995 \le x_1 \le 0.970;\ 0{,}005 \le x_2 \le 0.020;\ 0.001 \le x_3 \le 0.010. \quad (2)$$

As output, parameters were chosen: y_1—tensile strength of threads; y_2—strength of the threads in the loop; y_3—the diameter of the growth retardation of culture microorganisms *St.aureus*.

3. RESULTS AND DISCUSSION

When developing the plan of experiment, factor space in the form of simplex is used; so, the created software is provided for the transition from ordinary Cartesian co-ordinates to a special simplex system. The points, which determine the relative content of each component, are postponed along the corresponding faces of the simplex. At the vertices of the simplex, each $x_i = 1$, and then, they are determined by level lines parallel to the opposite side of the simplex. For a three-component mixture, the simplex has a form of tri-

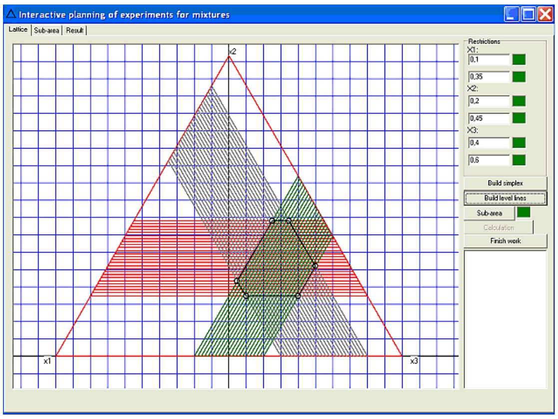


Fig. 1. Factor space for the experiment conducting.

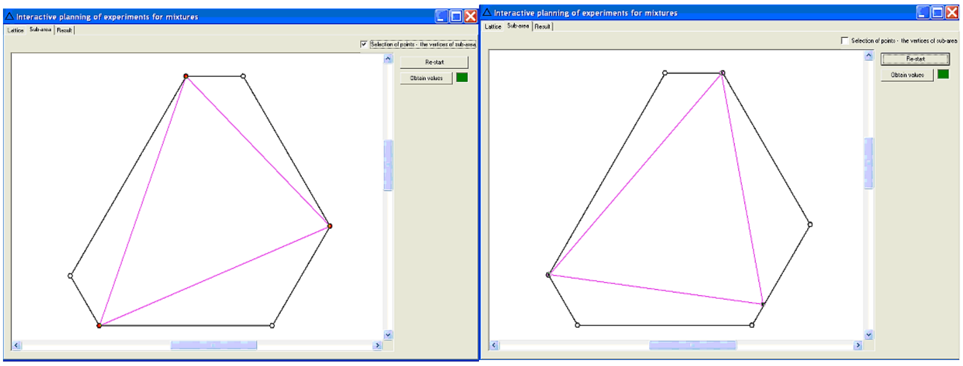
angle on the plane with vertices x_1, x_2, x_3 (Fig. 1). Each vertex of the simplex is an independent component of the mixture; the points contained on the side of the triangle correspond to the systems of the pairs of ingredients; the points in the middle of simplex correspond to the composition of the mixture of all three components. In particular, the value of the content of first component (x_1) at the vertex x_1 is equal to one, and values on the opposite side (x_2, x_3) are zero.

The content of ingredients in q-component mixture may vary from 0 to 1 or in the middle of this interval, which is determined by the requirements for the properties of the created compositions. In this case, it is necessary to investigate only the $(q-1)$-dimensional simplex subregion of the complete $(q-1)$-dimensional region. The subregion is defined by restrictions on the content of all q components. The developed program allows obtaining automatically the area for the experiment conducting for all possible combinations of the composition, including those with uneven content of components, when the concentration of one or two of them is less than other concentrations by several orders of magnitude. A limited area of irregular shape, which is the factor space for the experiment, is obtained by imposing restrictions on the concentration of ingredients. The factor space for compositions with a comparable content of components is shown in Fig. 1.

The planning of experiments by the simplex-lattice method is carried out in a subregion 'similar' to the original simplex; so, in a polygon, it is necessary to distinguish a subregion of triangular shape. This triangle, firstly, must lie completely inside the 'cut' area and, secondly, most fully cover it. The program allows us to construct interactively an area in the form of a triangle inside the found subregion. Defining the points of its vertices can be done in two modes: by selecting the option 'mode of selecting the points of the vertices of the subdomain' or by cancelling it. Figure 2 presents a uniformly enlarged factor space and examples of the desired subregion obtained using both modes.

Therefore, the next step is to select a subregion based on the empirical considerations of the researcher. Figure 3 shows a transformed subarea that can be used to build a work plan for experiments. The lower right field of figure shows the co-ordinates of the vertices of the triangle in the simplex system.

When planning an experiment in three-component systems, in which the content of one or two ingredients is several orders of



a *b*

Fig. 2. Uniformly enlarged factor space and examples of subareas obtained by different options: *a*) 'selection of points—the vertices of the subarea', *b*) without this option.

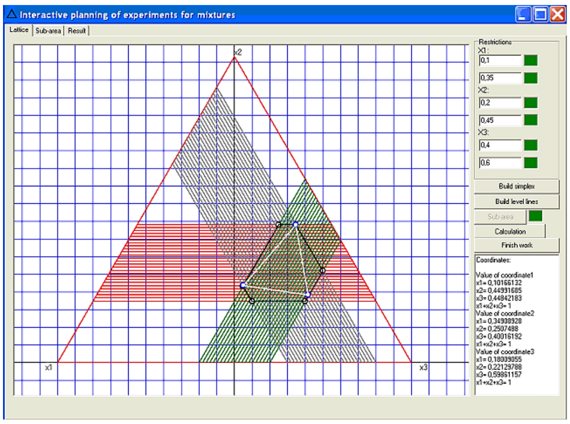


Fig. 3. Transformed subarea to build a work plan of experiment.

magnitude less than the third component, the factor space has an unusual appearance. It can be a strip, whose linear dimensions differ by hundreds or thousands of times, or a point region of very small size. In this case, a uniform increase in the ‘cut’ area and the interactive construction of the subregion become impossible. In the three-component composition studied in this work, the content of nanoparticles of alumina and carbon nanotubes is 500–1000 times less than the main component (polypropylene). The factor space obtained with the help of software has the form of a point, which is located near the vertex x_1 of the simplex (Fig. 4).

To solve this problem, in the program, special algorithms were implemented, which provide an uneven increase of the resulting ‘cut’ area with the mandatory preservation of the mathematical co-

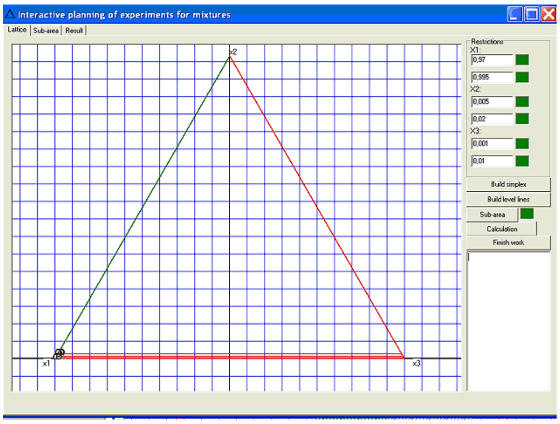


Fig. 4. Factor space of experiments for the studied system.

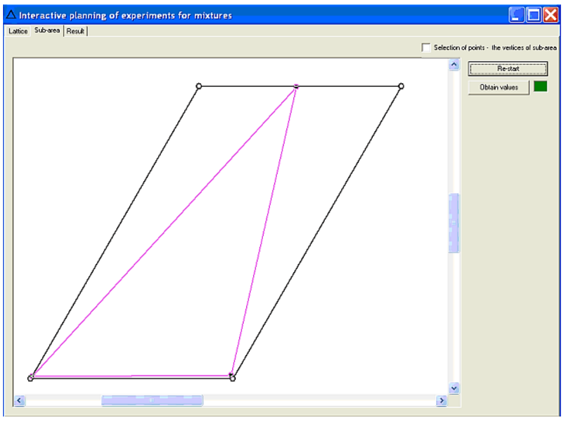


Fig. 5. An enlarged factor space with a triangular subarea.

ordinates (Fig. 5).

This is achieved automatically by entering the coefficient that changes the size of the subregion in a certain direction. Next, in an enlarged bounded area, we should choose a simplex in the form of a triangle, which will be the basis for creating a working plan of experiments.

The properties of three-component systems are most often described by algebraic polynomials. The program provides the construction of a plan of experiments, using, for choice, three types of equations: quadratic, incomplete cubic and cubic ones. As a response function that relates the output parameters to the values, which change during the experiments to describe the test mixture, we used a polynomial of incomplete third order:

$$\hat{y} = \beta_1 x_1 + \beta_2 x_2 + \beta_3 x_3 + \beta_{12} x_1 x_2 + \beta_{13} x_1 x_3 + \beta_{23} x_2 x_3 + \beta_{123} x_1 x_2 x_3 , \quad (2)$$

where β_i, β_{ij}, β_{ijk} are coefficients of the polynomial, and $i \neq j \neq k = 1$, 2, 3.

The working plan of the experiment was found, according to the standard plan for the incomplete cubic model in pseudo-co-ordinates (z) [16]. Calculations were performed, according the matrix ratio $X = AZ$, where the elements of the matrix A are the co-ordinates of the vertices of the constructed simplex subregion; X and Z are the matrices of the plan, namely, for the required working and for full simplex, respectively. In the program, the plan of experiment is obtained by pressing the button 'Calculation' in previous form (Fig. 6).

In order to establish the effect of the concentration of alumina nanoparticles and carbon nanotubes on the mechanical and bactericidal properties of polypropylene monofilaments, in accordance with the plan experiments, were performed, and the output parameters were determined (Table). Based on the data in this Table, with the help of previously developed software [18], we calculated the coefficients of the polynomials (2) by the least squares' method and obtained a set of Eqs. (3), which is a mathematical model of the studied process in z-co-ordinates:

$$\begin{cases} y_1 = 490z_1 + 550z_2 + 654z_3 + 280z_1z_2 + 210z_1z_3 - 90z_2z_3 + 2560z_1z_2z_3, \\ y_2 = 280z_1 + 320z_2 + 390z_3 + 240z_1z_2 + 260z_1z_3 + 140z_2z_3 + 1878z_1z_2z_3, \\ y_3 = 17.2z_1 + 20.9z_2 + 22.3z_3 + 2.6z_1z_2 + 18.2z_1z_3 + 19.2z_2z_3 + 134z_1z_2z_3. \end{cases} \quad (3)$$

The model was tested for adequacy, *i.e.*, the ability to predict the results of experiments in some areas with the required accuracy [19]. To do this, additional studies were performed at control

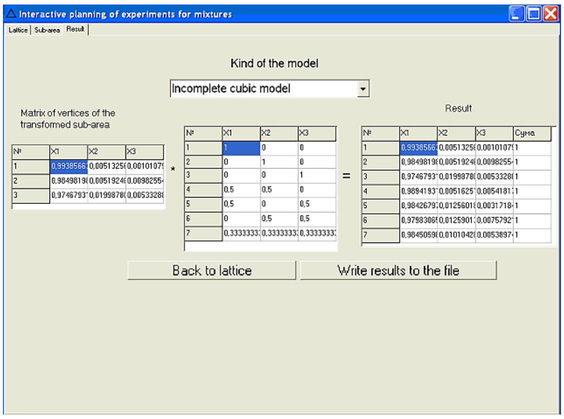


Fig. 6. The plan of experiment constructed by program.

TABLE. Influence of nanoadditive concentration on the properties of PP monothreads.

Output variable	Experiment number						
	1	2	3	4	5	6	7
y_1	490	550	645	590	620	575	700
y_2	280	320	390	360	400	390	470
y_3	17.2	20.9	22.3	19.7	24,3	26.4	29.5

points, and the prediction error of the output variable (ξ) was calculated, depending on its co-ordinates at the simplex. All the determined values of ξ were less than 1, *i.e.*, the control points were chosen optimally. Student's criterion (t_p) was also calculated for all the output variables and compared with tabular data. Determined on the basis of experimental and tabular data, the values of t_p are ranged from 0.033 to 1.659 for all studied initial parameters. This indicates that the developed model is adequate.

The ratio of components in the studied composition to obtain PP threads with a given set of characteristics was determined, using the generalized desirability function proposed by Harrington to optimize processes with a large number of responses [20]. To calculate the value of the desirability criterion, the determined values of the responses (y) were converted into the dimensionless desirability scale for each output parameter. Partial desirability functions were determined, using exponential dependence, pre-setting the worst and best values for each variable. The generalized value of the Harrington criterion was calculated as the geometric mean of the partial desirability functions. Using the generalizing function for multicriteria search of the optimal composition of the PP/Al_2O_3/CNT mixture, with the help of previously developed software [18], the content of the initial components of the mixture in z-co-ordinates was calculated. Then, using the matrix equation shown above (Fig. 6), we went to the working x-system. The criterion of desirability was equal to 0.9844, and the optimal concentrations of the ingredients of the mixture were as follow [wt.%]: polypropylene—of 98.35; alumina nanoparticles—of 1.11; carbon nanotubes—of 0.54. The compromise values of the responses, which characterize the quality of the modified threads, are as follow: tensile strength and loop strength—of 701 and 473 MPa, respectively, and the diameter of the growth retardation zone of *St.aureys* ATCC 25923—of 29.8 mm.

Studies of the properties of nanofilled monothreads formed from the mixture of optimal composition showed that the introduction of two nanoadditives Al_2O_3/CNT in the ratio of 1.1 wt.%:0.5 wt.% allows to increase their tensile strength by 1.7 times, compared with the best samples of polypropylene textile threads. The elasticity of the modified threads also increases; the strength in the loop is of 68% of the tensile strength. They show good manipulative properties, and they fix the surgical node well. The introduced nanoparticles do not impair the monolithic nature of the threads, and their surface remains smooth. The modifying additives also provide threads with high antimicrobial and antifungal activity to all investigated test-strains of microorganisms and fungi. The diameters of their growth retardation (D) around the holes are in the range 21.7–31.4 mm, which, according to the method used [10], is even better than norm, which is $15 < D < 25$.

4. CONCLUSIONS

In Builder environment in C++, software has been developed that allows in interactive mode to build the plan of experiment for various three-component mixtures, using three types of models of dependence of output parameters on the content of components—quadratic, incomplete cubic and cubic ones. The program allows us to solve the problem of planning an experiment for mixed compositions, in which the content of one or two components differs from the others by hundreds and thousands of times. This is achieved due to an algorithm developed in the software, which provides an uneven increase of the area in factor space with the mandatory preservation of the mathematical co-ordinates.

With the help of the developed software, the plan of experiments is constructed, and the mathematical model for the investigation of three-component system ‘polypropylene/alumina/carbon nanotubes’ is created. As shown, the model makes it possible to predict the results of investigation in a given area with the required accuracy, *i.e.*, it is adequate. Using the generalizing Harrington's criterion, the composition of the test mixture was optimized as follows: PP—of 98.35, Al_2O_3—of 1.11, CNT—of 0.54 (in [wt.%]).

As established, the use of aluminium-oxide nanoparticles and carbon nanotubes as modifiers in a ratio of 1.1 wt.%:0.5 wt.% provides polypropylene threads with high antimicrobial and antimycotic activity to all WHO-recommended test-strains of microorganisms and fungi. At the same time, it increases the strength of PP threads at break and in the loop by 1.7 and 1.9 times, respectively. Due to the low content of nanoadditives, the toxicity and the cost of modified threads are minimal; so, they can be successfully used in medicine as surgical suture materials, mesh grafts in reconstructive surgery, as well as raw materials for the production of special protective suits.

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