Preliminary Optimization of Composite Compositions Based on Modified Sosnowsky's *Heracleum*

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Composite materials based on modified stems of *Heracleum sosnowskyi* (Sosnowsky's hogweed) and polyurethane binder are used for thermal insulation of building structures. The purpose of this study was to create a mathematical model for the optimization of composite compositions and the prediction of their properties. Numerical methods of mathematical statistics were used and nomogram plots were obtained. It was possible to select optimal compositions for the given characteristics of composites based on modified stems of *H. sosnowskyi* and polyurethane binder and predict the thermophysical properties of composites by knowing their composition. To produce thermal insulation boards with a thermal conductivity coefficient of 0.05 W/(m°C) it was necessary to use particles of *H. sosnowskyi* with a size of approximately 5 mm. The ratio of plant raw material and polyurethane binder was approximately 3:1 by weight. The bending strength of the thermal insulation boards was 1.56 MPa, and the compressive strength was 0.27 MPa.

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Keywords: Sosnowsky's Heracleum; Grinding; Modification; Analytical optimization; Digital technologies; Material strength; Thermal conductivity

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INTRODUCTION

Thermal insulation materials based on porous polymers are one of the main types of thermal insulation used in the insulation systems of building structures. For synthetic building materials, foamed plastics and materials based on natural fibers have flammability and biological resistance issues, including resistance to decay (Umnyakova 2017; Ter-Zakaryan *et al.* 2021). The presence of water vapor suggests the possibility of moisture condensation on the internal surfaces of pores and capillaries, which can provoke the biological destruction of the material forming the matrix (Ter-Zakaryan *et al.* 2022; Zhukov *et al.* 2022). Improving fire and bio-resistance is increasingly of concern for thermal insulation materials based on natural fibers, including cellulose fibers (Gnip *et al.* 2005; Stepina *et al.* 2023).

There are two main reasons organic thermal insulation materials catch fire and break down: (1) the properties of the matrix substance, and (2) the presence of oxygen in the porous structure as the main part of the vapor-air mixture around the material and inside its pores. Oxygen, as an oxidizing agent, accelerates the processes of putrefaction and, in the case of fire exposure, creates favorable conditions for combustion (Zhukov *et al.* 2014; Efimov *et al.* 2020).

Wood fiber and its cellulose fibers can be changed in several ways. Surface treatment is thought to be the most technologically advanced and fastest method, but the best results are obtained by using modifiers that chemically react with hydroxylated substrates to create an organic matrix of materials (Pokrovskaya 2009; Koteneva *et al.* 2011).

Taking into account that cellulose is the main component of natural fibers, its modification allows changing the properties of the initial material in the direction of increasing its bio- and fire resistance (Koteneva *et al.* 2009). Koteneva and Kotlyarova (2010) and Stepina (2018) investigated the possibility of chemical interaction under conditions of "soft" modification using boron-nitrogen compounds as modifiers. The object of the research described in the article was thermal insulating materials (TIM) based on *Heracleum sosnowskyi* (Sosnowsky's hogweed) and polyurethane binder. The appearance of the samples of the received TIM of different granulometric compositions is shown in Fig. 1. Modification of plant raw materials has some positive effect on the appearance of samples. After modification, the dark gray deposit was eliminated by the impact of mold fungi on the surface of the material. Composites show a more pronounced golden coloration (Sidorov *et al.* 2009; Stepina and Sodomon 2022). It was previously found that modification of *H. sosnowskyi* with boron-nitrogen compounds leads to an increase in thermostability of plant raw materials. The process of degradation of the main component – cellulose – occurs at a higher temperature, and the value of mass loss during burning of the substrate is significantly reduced (Stepina *et al.* 2024). In addition, modification with boron-nitrogen compounds increases the strength of composites based on *H. sosnowskyi* (Sodomon and Stepina 2022).

The durability of materials based on *H. sosnowskyi* depends on the operating conditions. It is safe to say that in accordance with previous studies (Stepina *et al.* 2022), the durability of composites based on *H. sosnowskyi* modified with boron-nitrogen compounds is higher compared to composites based on unmodified *H. sosnowskyi*. This is due to 100% biostability of composites based on modified *H. sosnowskyi* and their increased strength compared to composites based on unmodified *H. sosnowskyi*.

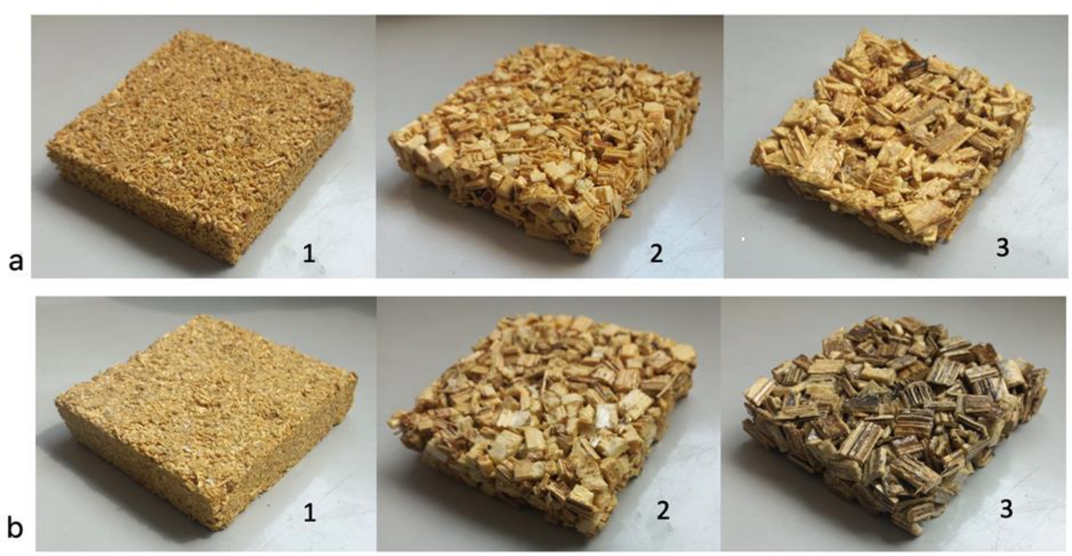

Fig. 1. Appearance of heat-insulating products: a, based on modified biostable raw materials; b, based on unmodified *H. sosnowskyi* at the size of its particles: 1 to 1 mm; 2 to 5 mm; 3 to 10 mm

Establishing the dependence of material properties on their component composition is one of the main tasks of building materials science, which is solved in modern conditions, including the use of digital methods (Popov *et al.* 2017; Pan *et al.* 2023). At the stage of experimentation, the best results are achieved by applying statistical methods. A lot of people use methods that use D-optimal orthogonal plans and matrices of full and fractional factor experiments with response surface methodology (RSM) for designing experiments and processing their results (Ali *et al.* 2023; Sharko *et al.* 2023; Yang *et al.* 2024). New technologies are being used to study and predict the properties of building materials (Chen *et al.* 2023; Mei *et al.* 2023). Different ideas can be tested with RSM, hybrid DNN-HHO (hybrid deep neural network-horse herd optimization), random forest (RF), and chaotic optimization (Satin Bowerbird) models, along with predictions and artificial neural networks. The combination of high-end digital technologies and full-scale experimental studies is of particular interest.

Based on modern methods of organizing experimental studies and processing their results, a comprehensive research methodology and prediction of the construction material properties depending on their formulations and technological parameters has been developed at Moscow State University of Civil Engineering. Using the "digital twin" method, the goal is to make models that accurately reflect real processes while reducing the number of material-intensive active experiments. The method also aims to find solutions to the direct and inverse problems of digital modeling and ways to check the accuracy of the results (Bobrova *et al.* 2019; Gudkov *et al.* 2019).

The article discusses research that creates better ways to choose the ingredients for a thermal insulating material using modified Sosnowsky's *Heracleum* as a starting point. Researchers also aimed to predict what properties the material would have by using modern digital tools and carrying out real-life experiments.

EXPERIMENTAL

H. sosnowskyi is an invasive species whose distribution range covers many regions. *H. sosnowskyi* is a large herbaceous plant more than a meter high, but in many places specimens up to 3 or even 4 to 5 meters high can be found. The stem is furrowed-ribbed, rough, partly tufted, purple or with purple spots, bearing very large triple- or pinnately-divided leaves, usually yellowish-green in color and 1.4 to 1.9 m long. The root system is tap root, the bulk of the roots are located in a layer of up to 30 cm, with individual roots reaching a depth of 2 meters. The attractiveness of this plant as a raw material to produce construction materials lies in its wide distribution in the European part of Russia, its rapid renewability, and, at the same time, the need to remove it from agrobiocenoses due to its expansion and displacement of other species. In addition, the stems of *H. sosnowskyi* have a highly porous structure and can serve as a basis for the creation of insulation boards. Thus, the crushed stems of *H. sosnowskyi* of different fractional compositions (from 1 to 10 mm) were used as raw material to create thermal insulation boards. The average density of thermal insulation panels based on modified *H. sosnowskyi* is 182 kg/m^3.

To ensure biostability and increase thermostability, crushed stems of *H. sosnowskyi* were modified by impregnation with a boron-nitrogen compound, monoethanolamine (NB)-trihydroxyborate (MEATHB). Impregnation was carried out at room temperature (25 °C) by immersion in the modifier with constant stirring. Plant raw materials were mechanically cleaned from impurities and air-dried to a constant weight. A 30% aqueous solution of MEATHB was used for impregnation, and the soaking time was 1 h. After impregnation, the plant raw material was dried to a constant weight at a temperature of 105 °C. The modified plant raw material was used to create thermal insulation boards using polyurethane binder.

The properties of composite materials are influenced by two main groups of factors that can be considered as variable and that can take part in the experiment: technological and formulation factors. Technological are factors that are not deterministic and can be varied in the course of the experiment. Firstly, these are factors related to the primary processing and grinding of borschyvik (intensity of work of shredding organs, loading factor of the shredder by material, *etc*.) Secondly, these are factors related to the modification of *H. sosnowskyi*: consumption of modifiers, temperature, concentration of the working solution, *etc*. Thirdly, these are factors related to the manufacture of products: molding speed and movement of forming belts, pressing force, temperature. The same group includes the formulation parameters and characteristics of the base raw material (*H. sosnowskyi*). It is these parameters that are accepted as variable factors and to the greatest extent meet the objectives of the research presented in the article.

Table 1. Experimental Conditions

Name of factor	Symbol X_i	Factor mean ($\underline{X}_i$)	Variation interval (ΔX_i)	Factor values at levels	
				-1	+1
Consumption of modified *H. sosnowskyi* (kg/m^3)	X_1	160	20	140	180
Particle size of modified *H. sosnowskyi* (mm)	X_2	4	2	2	10
Binder consumption (kg/m^3)	X_3	64	8	56	72

In the first stage of the experiment, the influence of all significant factors (having statistical significance) on the performance characteristics of the material was evaluated, and the most significant factors were singled out (consumption of modified *Heracleum* (kg/m^3); particle size of modified *H. sosnowskyi* (mm); binder consumption (kg/m^3)). The dependence of thermal conductivity (Y_1), flexural strength (Y_2), and compressive strength (Y_3) on the varying factors was taken as a response function. The experimental conditions are presented in Table 1. Factors in the table do not sum up to 100 percent, because they are given in units of flow rate (kg/m^3) and length of borer particles (mm). The factors are non-interdependent, so one of the basic requirements of mathematical planning is met, and it is possible to investigate the influence of varying factors on the result.

The experiment was conducted under the conditions shown in Table 1, using the full three-factor D-optimal planning matrix, as well as processing these results afterward in the Statistika program to obtain regression equations. The confidence intervals for equation coefficients were Δb_1 = 0.005 W/(m °C) for thermal conductivity, Δb_2 = 0.12 MPa for bending strength, and Δb_3 = 0.006 MPa for compressive strength. Coefficients that were not significant were set to 0, and the polynomials took on their final shape:

For thermal conductivity:

$$Y_1 = 0.059 + 0.022X_1 - 0.01X_2 + 0.0128X_3 + 0.014X_1 X_3 \quad (1)$$

For bending strength:

$$Y_2 = 1.09 - 0.32X_1 - 0.31X_2 + 0.24X_3 + 0.14X_2 X_3 - 0.16X_2 \quad (2)$$

For compressive strength:

$$Y_3 = 0.26 - 0.03X_1 - 0.02X_2 + 0.04X_3 + 0.01X_2X_3 \quad (3)$$

The analysis of response functions (algebraic polynomials) of polynomials 1 to 3 allows for the evaluation of the degree of influence of varying factors on the results. The amount of modified *H. sosnowskyi* and binder used primarily affects the product's thermal conductivity (Y_1, W/(m °C)). As the values of the factors go up, the thermal conductivity goes up (coefficients at X_1 and X_3 equal 0.02), but as the amount of modified *H. sosnowskyi* goes up, the thermal conductivity goes down (coefficient at X_2 equals -0.01). The pairwise interaction at X_2 and X_3 points to a possible synergistic effect of raising the amount of modified benzaldehyde and binder at the same time. The remaining coefficients of the polynomial were less than the confidence interval ($\Delta b_1 = 0.005$) and were assumed to be insignificant. Heat capacity does not directly affect the thermal characteristics of the building and characterizes the ability of the material to accumulate heat (when heated) and give it away when the temperature drops in the room or on the outer perimeter of the building. To the greatest extent, the thermal properties of the structure are assessed by thermal resistance, which in insulation systems with the use of insulation on the basis of *H. sosnowskyi* is in the range of 1.7 to 2.8 m^2 °C/W at the thickness of the insulation layer from 100 to 200 mm.

The bending strength of the products (Y_2, MPa) depends to the greatest extent on the size of the particles of borscht (coefficients at X_2 equal to -0.31 and at X_2 equal to -0.16). The decrease in bending strength with increasing particle sizes can be explained by the particle shape and nonlinear character. With small particle sizes, the strength increases, and with their increase, it begins to decrease. The determination of the optimum length is one of the analytical optimization problems.

An increase in the consumption of *Heracleum* determines a decrease in flexural strength (coefficient at X_1 equal to -0.32), while an increase in the consumption of polyurethane binder increases flexural strength (coefficient at X_3 equal to 0.24). The result of retaining more binder on larger borer particles explains the significance of the pairwise interaction (coefficients at X_2 and arX_3 are equal to 0.14). The other coefficients of the polynomial were less than the confidence interval ($\Delta b_2 = 0.12$ MPa), and were assumed to be insignificant and equal to 0.

The compressive strength of the products (Y_3, MPa) was determined to the greatest extent by the binder consumption and its properties (coefficient at X_3 equal to 0.04). The influence of *H. sosnowskyi* consumption and the size of its particles was also significant but to a lesser extent. There is a pairwise interaction (X_2, X_3) that is important to understand because of how the particles stick to the binder and how the *H. sosnowskyi* particles interact with the binder.

RESULTS AND DISCUSSION

Analytical optimization is based on the hypothesis that the obtained polynomials (1 to 3) are algebraic functions. In this case, they can be considered as functions of three variables (X_1, X_2, and X_3), using methods of mathematical analysis to obtain the values of partial derivatives and optimized functions. Analytical optimization has been used in solving technological problems and studying the properties of many types of building materials (Semenov *et al.* 2018; Zhukov *et al.* 2020; Lyapidevskaya *et al.* 2021).

The extremum of Eq. 2 was determined, differentiated by X_2, and equated to the partial derivative 0:

$$\frac{\partial Y_2}{\partial X_2} = -0.31 - 0.32X_2 + 0.14X_3 = 0$$

As a result, the following optimization function was obtained.

$$X_2 = -0.97 + 0.44\,X_3 \tag{4}$$

This value was substituted into Eqs. 1 to 3. The equations were solved with respect to the optimization function and the optimized functions were obtained (5, 6, 7):

$$Y_1 = 0.069 + 0.022X_1 + 0.018X_3 + 0.014X_1X_3 \tag{5}$$

$$Y_2 = 1.41 - 0.32X_1 + 0.47X_3 \tag{6}$$

$$Y_3 = 0.27 - 0.03X_1 + 0.12X_3 \tag{7}$$

To construct the nomogram, the obtained optimization functions (4 to 7) were translated into a graphical form and the individual graphs were combined into a single system. A nomogram for predicting the properties of products or selecting their formulation corresponding to the given properties is presented in Fig. 2. The nomogram was used for selection of composition and prediction of product properties.

The nomogram (Fig. 2) includes four sectors: I—to determine the thermal conductivity (W/(m °C)); II—to determine the flexural strength (MPa); III—to determine the compressive strength (MPa)and MPa; and IV—to determine the optimum particle length of the modified *H. sosnowskyi* (mm).

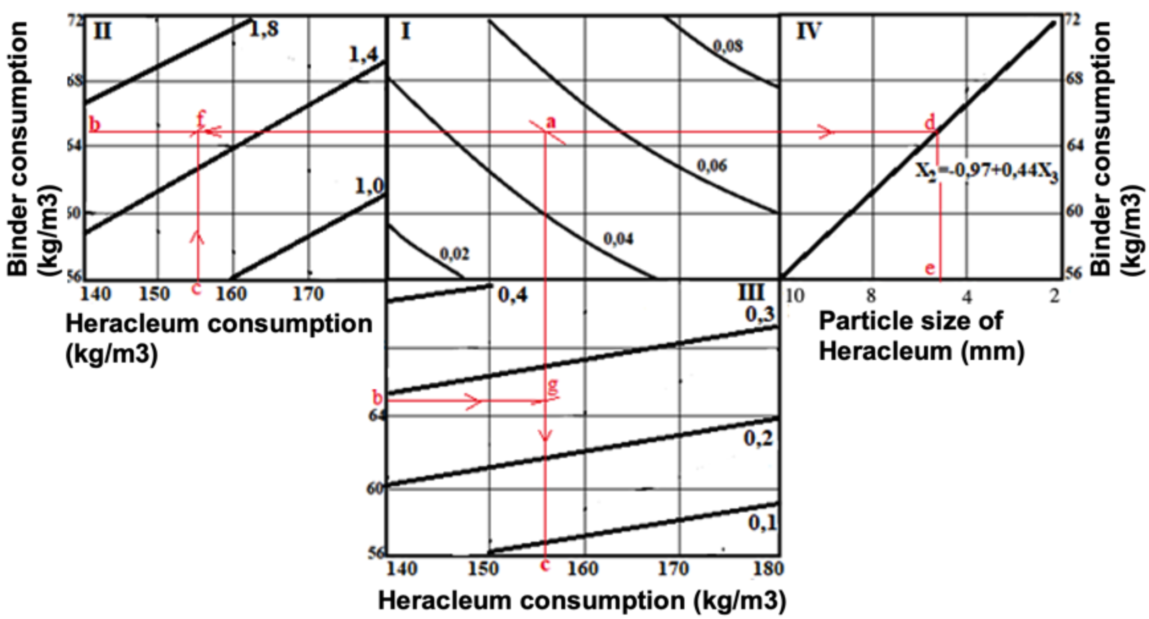

Fig. 2. Nomogram for composition selection and prediction of product properties. I—to determine the thermal conductivity (W/(m°C)); II—to determine the flexural strength (MPa); III—to determine the compressive strength (MPa) and MPa; and IV—to determine the optimum particle length of the modified *Heracleum* (mm)

To solve the inverse problem of determining the flow rate of components corresponding to the given characteristics, the desired thermal conductivity (point "a" in sector I) was set equal to 0.05 W/(m °C) and the perpendiculars on the axes were lowered to get the points "b", "c", and "d". The point "b" corresponds to the consumption of binder

65 kg/m^3, the point "c" corresponds to the consumption of modified *H. sosnowskyi* 156 kg/m^3, and the point "d" is obtained by the intersection of the perpendicular with the graph of the optimization equation. In sector II, the bending strength (for the binder and modified *H. sosnowskyi* consumption), was determined to be equal to 1.56 MPa (point "f"). In sector III, the compressive strength (for the binder and modified *H. sosnowskyi* flow rates) was determined to be equal to 0.27 MPa (point "g"). In sector IV, the optimum particle size of the modified *H. sosnowskyi* was determined to be equal to approximately 5 mm (point "e") (Fig. 2). The nomogram, presented in Fig. 2, is a summary of graphical dependencies of polynomials obtained as a result of analytical optimization of basic regression equations obtained as a result of statistical processing of experimental data.

The solution to the direct problem of predicting the properties of thermal insulation material depending on the accepted values of varying factors can also be carried out with the help of the developed nomogram and the implementation of a computer program. The program algorithm includes the following blocks: data input (values of varying factors in natural terms), data coding, calculation by polynomials (4 to 7), and the output of results to print and display (Fig. 3). The data are entered into the program in physical terms (Xin): consumption of modified *H. sosnowskyi* Cb (kg/m^3), particle size of modified borer Sb (mm), and consumption of binder CPU (kg/m^3). Coding of factors (bringing them to the interval [-1; +1]) is carried out using the data of Table 1 by the formula:

$$X_i = \frac{X_{ci} - X_{ni}}{\Delta X_i} \tag{8}$$

where X_{ci} is the average value of the i-th factor; ΔX_i is the variation interval of the i-th factor; and i is the factor number (1, 2, 3).

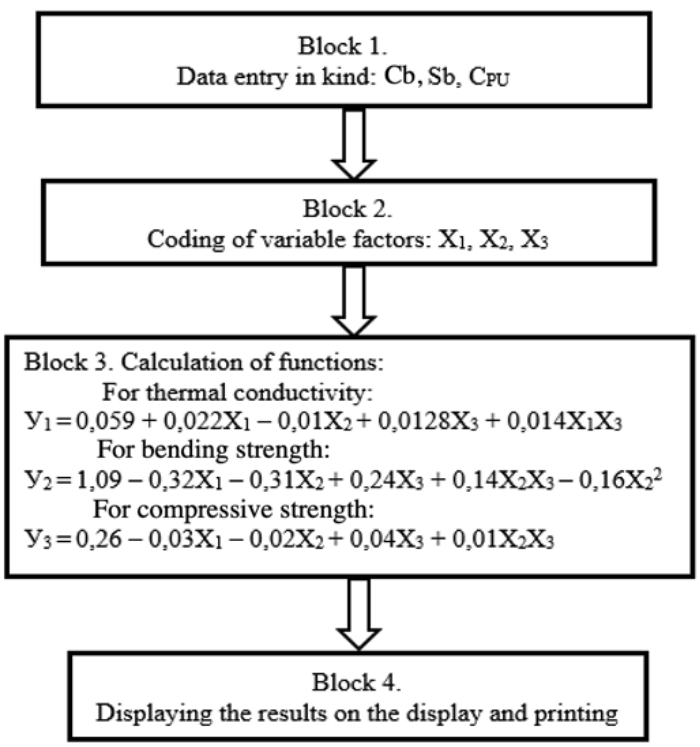

Fig. 3. Computer program algorithm

When using a nomogram or computer calculation to obtain the Dainvolveseck coefficients, it is important to ensure that the results are convergent. This is also true when completing a series of natural extra experiments and judging the properties of samples made with a certain flow rate of components. In the optimization and statistical experiment, the thermal conductivity of the material made from *H. sosnowskyi* and polyurethane binder was investigated. The difference between theoretical and experimental data on thermal conductivity was no more than 7.9%, which was the same level of accuracy used for the measurements.

CONCLUSIONS

1. Using numerical methods of mathematical statistics, nomogram plots were obtained, with the help of which it was possible to select optimal compositions for the given characteristics of composites based on modified stems of *H. sosnowskyi* and polyurethane binder. *Vice versa*, it was possible to predict the thermophysical properties of composites by knowing their composition.
2. Thermal insulation material based on modified *H. sosnowskyi* fibers and synthetic binder was obtained using the described methods. The obtained dependences made it possible to evaluate the influence of varying factors on the result, and the interpretation of dependences allowed solution of the prognostic problem of predicting the properties of products.
3. To produce thermal insulation boards with a thermal conductivity coefficient of 0.05 W/(m°C) it was necessary to use particles of *Heracleum* with a size of about 5 mm. The ratio of plant raw material and polyurethane binder was approximately 3:1 by weight. The bending strength of the thermal insulation boards was 1.56 MPa, and the compressive strength was 0.27 MPa.

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