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DESIGN OF BIOCOMPOSITES BASED ON A GLUTINOUS MATRIX WITH A COMBINED CONTENT OF WOOD FLOUR AND CHOPPED STALKS OF GRAIN CROPS

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This study considers biocomposite materials based on a glutinous matrix and fillers of plant origin. Waste from the woodworking industry and the agricultural sector is a promising raw material for obtaining fillers for biocomposite materials, which are characterized by a high degree of environmental safety and renewable raw material base.

The task addressed is to optimize the amount of technological additive (water) in the glutinous composition, which contributes to the formation of a dense structure of the biocomposite material with a compact arrangement of particles of the combined mixture of fillers.

In the process of research, the effectiveness of using fillers of different granulometric composition against the effect of mechanical loads was determined. The complex effect of fillers of plant origin on the mechanical characteristics of biocomposite materials was studied, which made it possible to define the optimal composition of the biocomposite.

An analysis of the effect of the amount of moisture in the composition on the formation of the structure and the resistance of the biocomposite material to the effect of static and dynamic loads was carried out. The maximum values of the ultimate compressive strength (115–120 MPa) were obtained for biocomposites containing a mixture of fillers of different granulometric composition (40–80% of cereal stalk particles, the remaining particles of wood flour) provided that 30% of moisture is removed from the composition. The maximum impact strength (13.8 kJ/m²) was established for biocomposites containing 100% of crushed cereal stalks provided that 10% of moisture is removed from the composition.

The designed materials could be used to manufacture packaging elements that are disposed of after operation by recycling or through safe decomposition at landfills without harm to the environment

Keywords: biopolymer, plant fillers, moisture content, compressive strength, impact strength

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1. Introduction

Polymer composite materials based on synthetic matrices and inorganic fibers, in particular carbon and aramid [1], are widely used in the automotive, aircraft, electronics, construction, military, food, and textile industries, as well as sports industries, because of their high specific strength, corrosion resistance, and manufacturability. However, a number of significant problems arise related to the deterioration of environmental safety and the limited resource of the raw material base. This leads to the search for alternative sources of raw materials that are safe for the environment.

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Work [2] shows that helmets and sports products made on the basis of polymer composites containing synthetic (carbon fiber) and natural (coconut fiber) fibers have sufficiently high mechanical properties. Studies on polymer composites with a combined content of coconut and glass fibers [3] have also been conducted, which make it possible to partially improve the environmental friendliness of products. Therefore, one of the options for solving the problem is to replace synthetic fibers with fibers of plant origin (banana peel fibers, kenaf, technical hemp and sisal) [4].

In [5], it was established that biocomposites with high physical, mechanical, and tribotechnical characteristics can be obtained using banana peel fibers. Of the natural fillers,

special attention is paid to kenaf because of the combination of strength and environmental friendliness [6]. These fibers are safe at processing and do not form harmful substances during the disposal of biocomposite products based on them. This provides a high degree of biodegradability and recyclability, a reduction in the weight of products, an increase in the service life of technological equipment, as well as satisfactory mechanical properties [7, 8].

It should be taken into account that plant fibers in cross-section do not have a rounded shape but are similar to polyhedra, which causes variability in the mechanical properties of biocomposite materials. It is also necessary to take into account the high hydrophilicity of the fillers, which complicates the interaction with the polymer matrix [8]. Usually, design of biocomposite products involves replacing one of the components of the polymer composite material while the polymer matrix remains [9, 10] synthetic (epoxy, polyester resins). The use of thermosetting plastics as a matrix provides high resistance to atmospheric factors but reduces the biodegradability of the material and complicates the processing and disposal of biocomposite products.

In work [11], a polypropylene matrix was studied, which includes lignin and micro cellulose. The compatibility of the components was increased as a result of chemical treatment of the components and the use of talc. A positive result was obtained for composite materials containing lignin as there was an increase in Young's modulus, as well as tensile and bending strength. The authors showed the effectiveness of introducing lignin into the composition of the designed polymer composites as there was an increase in mechanical characteristics and biodegradability. The use of thermoplastic matrices provides processing of polymer composite products compared to matrices based on thermosets, but they are not environmentally friendly.

The use of thermoplastic matrices in biocomposites reduces their biodegradability and requires the use of raw materials of mineral origin, which are non-renewable. Therefore, it is advisable to replace synthetic polymer matrices with materials of natural origin with high biodegradability. This requires experimental studies on the compatibility of components and the features of structure formation, as well as determining mechanical characteristics of the designed biocomposite materials. These aspects predetermine the high relevance of this scientific area.

2. Literature review and problem statement

The use of biocomposites based on a synthetic matrix with a combined content of fillers of natural origin is widespread [11]. In this case, a synergistic effect is achieved, which contributes to the improvement of the mechanical characteristics of the material. At the same time, the use of microfillers complicates the technological processing of the composite. The application of fibers of natural origin increases the manufacturability and environmental safety of production [12]; however, the mechanical characteristics of the material decrease. In [13], it is noted that the use of natural fibers improves economic efficiency but, with an increase in their content, brittle fracture of the biocomposite occurs because of the fibers pulled out of the matrix. With an increase in the content of glass fibers, tensile and bending strength, elastic modulus and impact strength increase [14]. This indicates the feasibility of combining two types of fibers but, at the same time, the biodegradability of the material decreases.

Organic fibers of plant origin have high tensile strength and elastic modulus, which makes it possible to increase the bending strength and impact toughness of biocomposite materials [15]. Classic safety helmets are made using aramid fibers; therefore, to improve the impact toughness, the filler content is increased. This leads to an increase in the mass of the helmet, which worsens the ergonomic properties of the product. Bamboo fibers were used as a filler to form safety helmets based on urea-formaldehyde resin. As a result of impregnation with a synthetic matrix of the filler, the crystallinity of parts of bamboo stems increases, which improves the mechanical properties of the biocomposite material. This makes it possible to obtain products of lower weight and cost with increased resistance to dynamic loads, which are adapted to the effects of low and high ambient temperatures and humidity. Bamboo stems consist of cellulose, hemicellulose, and lignin, which ensures the biodegradability of the material in the natural environment and reduces the level of pollution. In [16], coconut shell was used as a filler for the nylon matrix. This allowed the authors to reduce the degree of plastic deformation, increase the shear strength, improve durability, and reduce the weight of the biocomposite helmet due to the uniform distribution of stresses in local volumes of a homogeneous structure.

An important factor affecting the mechanical properties of polymer composite materials is the morphology of the filler particles, in particular their size, which determines the strength and impact toughness of the material [17, 18]. These papers investigated the morphology and particle size distribution of filler particles, but the studies mainly concern mineral materials, which complicates the application of the results to the design of biocomposites with fillers of natural origin. In [19] it is noted that the use of spherical particles has an advantage over short rod-shaped particles since it increases resistance to static loads but, at the same time, reduces the impact toughness of the polymer composite material.

In [20], the effect of spherical aluminum oxide particles on the friction and wear resistance of polymer composites was investigated, which confirms the role of filler morphology in the formation of their mechanical properties. At the same time, due to the low filler content, the results do not allow for a full assessment of the regularities of the formation of highly filled systems. In [21], the influence of the morphology of filler particles on the strength and durability of materials was also investigated and confirmed, although this aspect was not given enough attention. The use of crushed pine needles [22] enables the formation of a biocomposite material with low density and cost, acceptable specific strength, recyclability and biodegradability. At the same time, such a material is characterized by lower adhesive strength, fatigue strength and durability compared to polymer composites containing synthetic fibers. It was established that biocomposites based on urea-formaldehyde resin [23] with the content of spherical particles are characterized by the highest tensile strength, compressive strength and wear resistance compared to materials reinforced with short or long fibers. At the same time, the high content of natural fillers helps increase the biodegradability of products, but the use of a synthetic matrix reduces their environmental friendliness.

The use of powdered filler, which is obtained by grinding hazelnut or peanut shells, for the formation of biocomposite materials based on low-density polyethylene leads to an increase in tensile strength and impact strength [24]. The increase in characteristics occurs in the case of a high degree of

homogenization of the composition, which is achieved after three or four stages of dispersion of the mixture. It was established that the optimal filler content is 40 wt. parts, which is explained by the ability of the binder to form chemical bonds between the filler particles. In [25], the effect of the content of the modifying additive on the compressive strength of biocomposites structured at different temperatures was investigated. It was established that a modifying additive with an optimal content is able to increase the intensity of structuring of the biopolymer binder. However, the use of additional treatments and modifying additives complicates the technological process of manufacturing products.

The use of synthetic components complicates the technological processing of the composition, reduces biodegradability, and could lead to an increase in the mass of polymer composite products, it reduces the service life of equipment as well. Therefore, in world practice, materials of natural origin are actively introduced as a reinforcing phase of biocomposite materials based on synthetic polymers or biopolymers, which makes it possible to expand the raw material base and increase environmental safety. At the same time, the introduction of fillers of natural origin into the composition reduces the adhesive strength, fatigue strength, impact strength and durability of biocomposite products, which requires investigating their structure and mechanical properties.

3. The aim and objectives of the study

The aim of our work is to design the composition of biocomposite materials based on natural components with different degrees of drying of fillers, as well as to determine their mechanical characteristics and structure. This will increase the resistance of glutinous biocomposite material with a combined content of natural fillers to static and dynamic loads.

To achieve this aim, the following objectives were accomplished:

- to determine the compressive strength of biocomposite materials at the optimal filler content;
- to analyze the fracture fractograms of biocomposite materials;
- to analyze the nature of the destruction of biocomposite samples as a result of applying a static load;
- to determine the impact strength of biocomposite materials at the optimal filler content.

4. The study materials and methods

The object of our study is biocomposite materials based on a glutinous matrix and fillers of plant origin. The subject of the study is the processes of forming the structure of biocomposite materials based on a composition containing a glutinous binder and a complex content of fillers of natural origin.

During the study, a hypothesis was put forward that removing excess moisture from the composition before pressing by forming biocomposite materials in a mold could enable compaction of the material and reduce the number of structural defects. This would improve the interfacial interaction between the components and increase the compressive strength and impact toughness of biocomposite materials.

The following assumptions and simplifications were adopted in the study:

- wood flour was dried at a temperature of 100–110°C for 40–50 min;
- the composition was compressed with a specific load of 130 MPa;
- heat treatment of the press composite was carried out for 90 min at a temperature of 140°C.

Biocomposite materials were formed on the basis of a glutinous matrix and a set of natural fillers (wood flour and crushed stalks of cereal crops). A 60% glutinous solution was obtained by mixing bone glue granules with water in a ratio of 60:40. The mixture in a sealed container was placed in a chamber of the drying cabinet SNOL 75/400 with a fan and kept for 2–3 h at a temperature of 50–55°C. The solution was periodically stirred to ensure homogeneity and to be used immediately after preparation. Before use, the wood flour was dried at a temperature of 100–110°C for 40–50 min to remove moisture, and sieved using metal mesh sieves (ISO 3310-1:2017) to obtain a powder with a particle size of 0.7 mm. The grinding of cereal stalks was carried out using a high-speed GoodFood PG1000 hammer mill with a working element rotation frequency of 20,000 rpm. The resulting mixture of crushed stalks was similarly dried at a temperature of 100–110°C for 40–50 min and sieved to obtain particles with a length of 3.5–4.0 mm. The dosage of components was carried out by weight using the analytical laboratory scales WPS 110/C/1 of the third accuracy class. The ratio of components was determined by the total amount of combined filler content, which corresponds to 100 wt. parts per 100 wt. parts of glutinous matrix.

The next stage involved mixing the components mechanically in the chamber of the laboratory mixer LZMK-1 for 10 s in order to avoid heating the composition and additional grinding of the components. The formation of samples of a given shape and size was carried out using a mold designed at the laboratory, consisting of a cylindrical matrix and punches. The composition was placed in the mold matrix and compressed with a specific load of 130 MPa. The mold with fixed punches was placed in the chamber of the drying chamber and thermally treated for 90 min at a temperature of 140°C. After cooling the mold, the sample was removed and tested.

Compressive strength was determined according to the ASTM D695 methodology [26], which involves the use of cylindrical specimens (height 30 mm and diameter 20 mm). The specimens are subjected to compression at a relatively low uniform loading rate (2 mm/min). Impact strength was determined according to the ISO 179 methodology [27]. The method involves applying a dynamic load to a biocomposite specimen, which is located on two supports of a pendulum impactor. The measurements imply registering the energy that the material absorbs before failure. The study was conducted on biocomposite specimens with a square cross section of 10 × 10 mm and a length of 60 mm.

5. Results of investigating the mechanical properties of biocomposite materials

5.1. Determining the compressive strength of biocomposite materials at the optimal filler content

The compressive strength of biocomposite materials containing 100% wood flour is 5.6 MPa (Fig. 1). Biocomposite samples formed without prior heat treatment of the composition in a thermal field have an increased moisture content in the biocomposite material at the forming stage. With an increase in the content of crushed cereal stalks, the total filler content

relative to the volume of the biopolymer matrix does not change, but there is a proportional decrease in the content of wood flour relative to the content of cereal stalks. Changing the ratio of the content of the solid phase components has little effect on the compressive strength of the biocomposite material, since with an increase in the content of crushed cereal stalks, the change in the characteristic occurs within 1.4–1.6 MPa.

The maximum values of compressive strength (115–120 MPa) were obtained as a result of mixing fillers of different granulometric composition (40–80% of grain stalk particles, the rest of the wood flour particles) provided that 30% of moisture was removed.

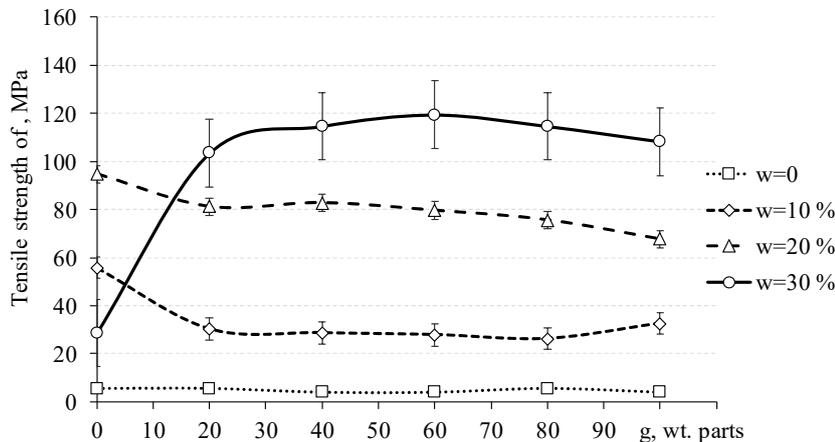


Fig. 1. Effect of the content of crushed cereal stalks on the tensile strength of biocomposite materials depending on the degree of drying of the composition (*W*)

Removing 10% of moisture from the composition at the preparatory stage of forming biocomposite materials leads to a 10-fold increase in the compressive strength of biocomposite materials containing 100% wood flour. This is due to the formation of a biocomposite material structure with fewer defects in the form of cavities and pores, the formation of which occurs as a result of intensive moisture removal during heat treatment at a temperature of 140°C.

The compressive strength of biocomposite materials whose compositions have lost 20% of moisture and are filled with wood flour particles is 94–96 MPa. The strength of such materials after processing the composition in a thermal field exceeds the strength by 93–96% of similar materials whose compositions have an increased moisture content in the composition. This is due to the formation of a dense structure of the biocomposite material due to the approximation of glutinous macromolecules and the formation of bonds between the active groups of the glutinous matrix and reactive groups on the surface of the filler.

5. 2. Analysis of fractograms of biocomposite materials

As a result of holding the composition containing wood flour in a thermal field at a temperature of 50–55°C for 20–25 min, moisture loss occurs in the amount of 10% by weight from the initial value. In this case, the compressive strength of the biocomposite material increases 10 times and is 55.7 MPa. Removing part of the moisture

at the stage of heat treatment of the composition enables the formation of a structure of the biocomposite material with fewer defects (Fig. 2, *a*). This is due to the transition of moisture to a gaseous state during the formation of biocomposite products at a temperature of 140°C. With increasing temperature, water molecules are intensively removed, which leads to an increase in the stress state, destruction of interfacial bonds, and the appearance of microscopic cracks (150–200 μm), cavities (80–100 μm) or pores (30–40 μm) (Fig. 2, *b*).

With an increase in the content of crushed cereal stalks to 20%, the content of wood flour decreases to 80%, which leads to a decrease in the compressive strength by 45%. In the case of an increase in the content of crushed cereal stalks to 80%, the compressive strength decreases by 52–54% compared to the strength of the biocomposite material containing a filler in the form of wood flour particles. The decrease in the strength of biocomposite materials is associated with an increase in the content of stalk particles, which have larger sizes compared to the particle sizes, so their surface energy is significantly lower. Under such conditions, a smaller number of chemical bonds are formed in the biocomposite material between the filler and the glutinous matrix. Also, the surface of cereal stalks has less roughness (Fig. 3, *a*) compared to the developed surface of wood flour particles (Fig. 3, *b*), which reduces the area of interfacial contact and the ability to form bonds.

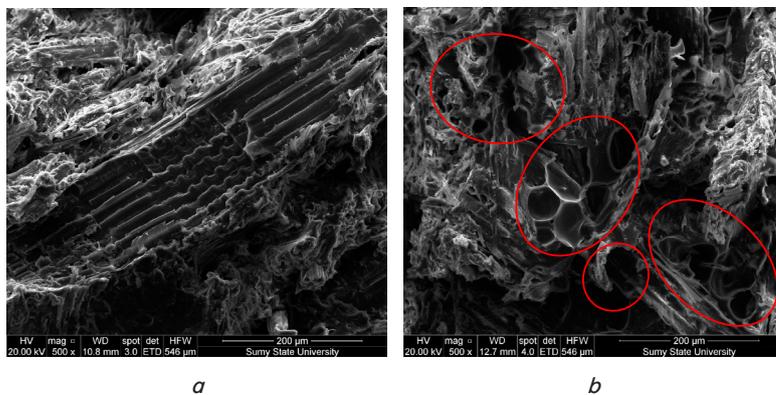


Fig. 2. Fracture fractograms of biocomposite materials with a degree of moisture loss: *a* – 10%; *b* – without moisture removal

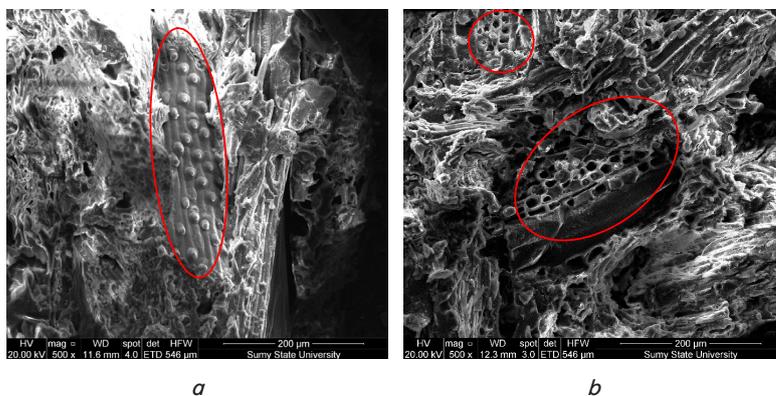


Fig. 3. Fracture fractograms of biocomposite materials filled with: *a* – particles of cereal stalks; *b* – wood flour

With an increase in the content of crushed cereal stalks, the compressive strength decreases to 75.4–81.2 MPa in the filling range of 20–80%. In the case of filling with stalk particles in an amount of 100%, the compressive strength decreases to 67.7 MPa, which is 28–30% lower compared to the strength of biocomposites, the compositions of which have lost 20% of moisture and contain 100% wood flour. The uniform decrease in this characteristic is associated with a gradual increase in the content of cereal stalk particles, which are randomly arranged in the glutinous matrix (Fig. 4, *a*) compared to the compact arrangement of wood flour particles (Fig. 4, *b*).

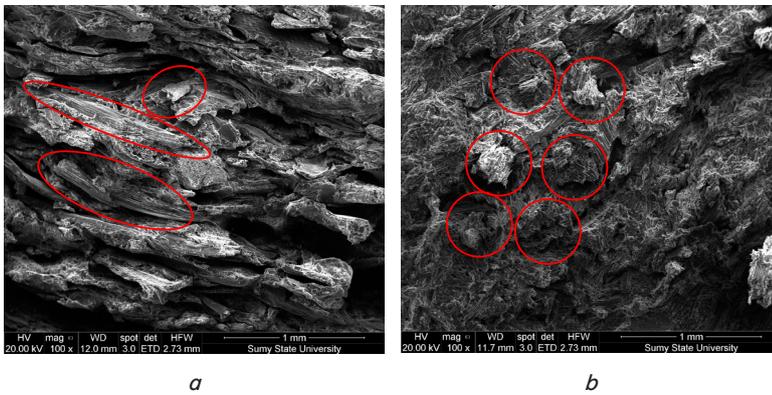


Fig. 4. Fracture fractograms of biocomposite materials: *a* – chaotic arrangement of grain stalk particles; *b* – uniform arrangement of wood flour particles

It can be argued that under the influence of static loading, an uneven distribution of stresses occurs in local areas of the biocomposite material, which causes cracks to appear in places of stress concentration.

5. 3. Analyzing the nature of destruction of biocomposite samples as a result of applying a static load

A biocomposite sample containing 100% crushed stalks of cereal crops is deformed similarly to a sample containing 100% wood flour. Under the influence of static load, the upper part of the sample is pressed into the lower part with the formation of a thickening in the central part of the sample (Fig. 5, *a, b*). In this case, delamination of the components occurs along the outer side surface of the cylindrical sample with the formation of vertical main cracks as a result of the action of tangential stresses. The edges of the main crack of the biocomposite sample containing 100% crushed stalks of cereal crops are not even due to the chaotic arrangement of stalk particles in the biopolymer matrix, which complicates the rectilinear propagation of the crack and leads to the tearing of filler particles from the matrix.

The destruction of biocomposite samples containing 100% wood flour occurs as a result of the appearance of cracks in the lower part of the cylindrical sample (Fig. 6, *a*) under the influence of elastic deformation, which indicates increased resistance to static loading of the biopolymer matrix. The appearance of vertical cracks indicates a low degree of plastic deformation, which occurs under the influence of tangential stresses. Biocomposite samples filled with particles of cereal stalks plastically deform with the formation of a thickened area in the central part of the sample (Fig. 6, *b*) without the appearance of main cracks. This

indicates the increased strength of the biocomposite material, the composition of which contains 10% less moisture, compared to a similar sample, the composition of which has an increased moisture content. The appearance of plastic deformation is associated with the formation of walls of the glutinous matrix of increased thickness compared to biocomposites filled with wood flour. This leads to the displacement of local areas of the glutinous matrix without delamination of the components.

Under the influence of static loading, the biocomposite material containing 100% wood flour undergoes destruction with the formation of a main crack (Fig. 7, *a*) and small-volume chipped areas in the lower and upper parts of the sample. The crack is located at an angle of 40–45°, which indicates the presence of normal and tangential stresses that cause plastic and elastic deformation. The presence of one crack indicates the resistance of the biocomposite material to elastic deformation and the presence of brittle fracture due to the increase in the rigidity of the chains of macromolecules of the glutinous matrix. The destruction of the biocomposite material, the composition of which has lost 20% of moisture and contains 100% of grain stalk particles, occurs similarly to the formation of a main crack (Fig. 7, *b*). Such a crack changes the direction of movement due to the presence of stem particles that are located at an angle of 85–95° to the direction of movement, as a result of which the crack is characterized by surface curvature in local areas.

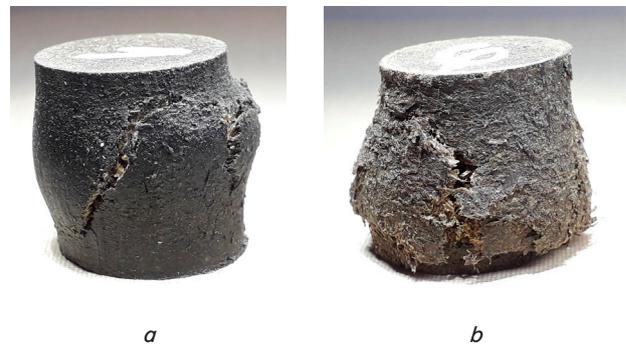


Fig. 5. General view of samples after compression failure with increased moisture content in the composition: *a* – 100% wood flour; *b* – 100% crushed cereal stalks

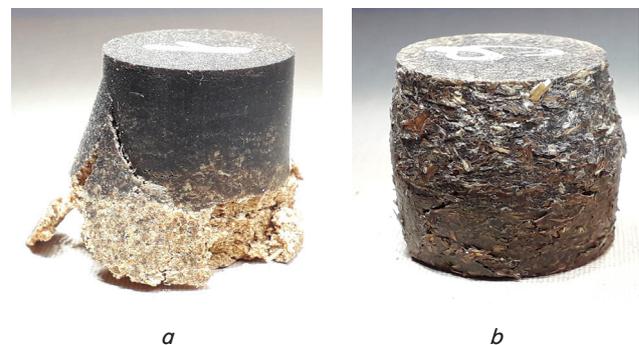


Fig. 6. General view of samples after compression failure with a moisture loss of 10% in the composition: *a* – 100% wood flour; *b* – 100% crushed cereal stalks

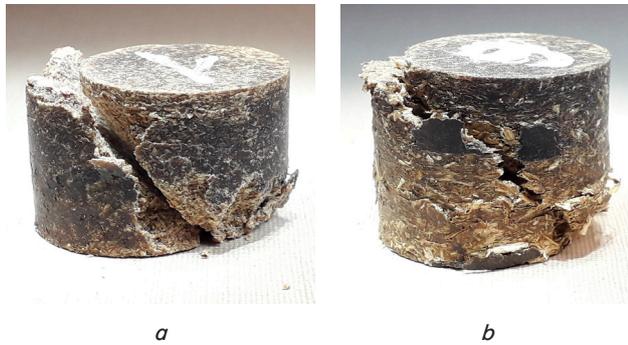


Fig. 7. General view of samples after compression failure with a moisture loss of 20% in the composition: *a* – 100% wood flour; *b* – 100% crushed cereal stalks

Biocomposite samples containing 100% wood flour and having lost 30% moisture in the composition fail as a result of delamination of the components in a plane located at an angle of 45° to the direction of the compressive force (Fig. 8, *a*). Failure of similar biocomposite samples containing 100% crushed cereal stalks occurs as a result of crack propagation at an angle of 60° to the direction of the static load (Fig. 8, *b*), which indicates the dominant influence of normal stresses.

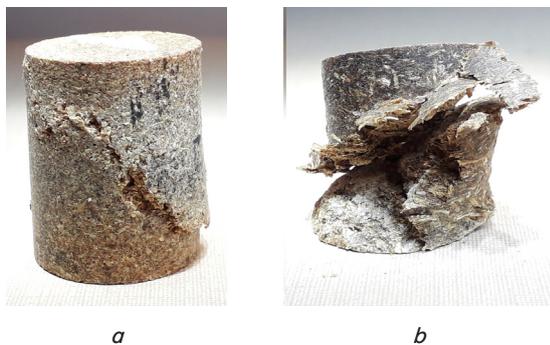


Fig. 8. General view of samples after compression failure with a moisture loss of 30% in the composition: *a* – 100% wood flour; *b* – 100% crushed cereal stalks

Therefore, under the influence of normal stresses, the biocomposite material undergoes elastic deformation, after which several main cracks form as a result of brittle fracture of the biocomposite.

5.4. Investigating the impact strength of biocomposite materials with optimal filler content

Biocomposites containing 100% crushed stalks of cereals have a 30% higher impact strength compared to biocomposites containing a mixture of fillers (40% wood flour and 60% crushed stalks of cereals). The stalk particles are larger and have an elongated shape and are also located perpendicular to the direction of dynamic loading, which complicates the process of spontaneous crack movement.

The maximum impact strength value (13.8 kJ/m^2) was obtained as a result of removing moisture in an amount of 10% from the composition containing 100% crushed stalks of cereals. The removal of moisture enables the formation of additional chemical bonds between the reactive groups of the glutinous matrix and the active groups on the surface of the filler particles.

Removing 20% moisture from the composition leads to a 16–18% decrease in the impact strength of biocomposite

materials compared to the impact strength of biocomposite materials whose compositions lost 10% moisture. This is due to an increase in the rigidity of the glutinous matrix macromolecules, which leads to a restriction of micro vibrations of the matrix macromolecule segments and reduces the material's ability to absorb the energy of dynamic loading.

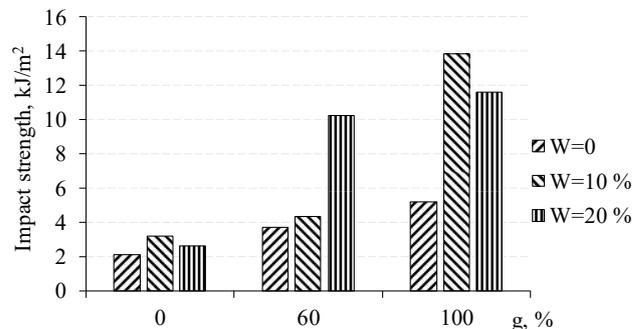


Fig. 9. Effect of the content of crushed cereal stalks on the impact strength of biocomposite materials depending on the degree of moisture removal in the composition (*W*)

6. Discussion of results based on investigating the mechanical properties and structure of biocomposite materials

Biocomposite materials based on a glutinous matrix contain water, which acts as a solvent for glutinous granules, however, with an increased moisture content in the composition, the compressive strength of such materials is within 4.0–5.6 MPa (Fig. 1). This is explained by the decrease in the resistance of the glutinous matrix to static loads since water molecules complicate the process of forming chemical bonds between the components of the biocomposite material [25].

The stiffness of the biopolymer matrix decreases due to the dominant influence of moisture contained in the composition and reduces the stiffness of the biopolymer matrix. Water provides dissolution of the glutinous binder, which after the formation of the biocomposite performs the function of a biopolymer matrix; however, with an increased moisture content, it has low mechanical characteristics. This leads to a decrease in the hardness and adhesive strength of the glutinous matrix, which causes plastic deformation in the central part of the biocomposite sample (Fig. 5, *a*). In this case, the stresses are distributed evenly due to the isotropy of the material, which contains spherical particles [18] of wood flour. The main crack is formed as a result of delamination of the components [2] of the biocomposite material due to the low adhesive strength of the glutinous matrix and is located at an angle of 45° to the axis of load action, which indicates the presence of normal stresses.

The compressive strength of a biocomposite material containing 100% grain stalk particles is 32.6 MPa (Fig. 1). In this composition, the compressive strength is 20% higher than the strength of biocomposites containing 80% grain stalk particles. With the same binder content in the system, the distance between the particles in the biocomposite material containing crushed grain stalks is greater (Fig. 4, *a*) compared to the distance between the spherical particles of wood flour (Fig. 4, *b*). In this case, the wall thickness of the glutinous matrix of the biocomposite material containing crushed stalks is greater. This provides an increase in the compressive

strength of biocomposite materials, since with a decrease in moisture in the composition, the rigidity of the framework of the glutinous matrix macromolecules increases, which determines the resistance of the matrix to static loads.

The compressive strength of biocomposites containing 100% wood flour is 94.8 MPa (Fig. 1) in the case of a 20% moisture loss of the composition. This is 40–42% higher compared to the strength of biocomposites of a similar composition with a 10% moisture loss in the composition and 93–96% higher compared to biocomposites whose compositions did not lose moisture. Removal of excess moisture ensures the compaction of the macromolecules of the glutinous matrix (Fig. 2, *a, b*), in the cells of which water molecules are located. In the case of macromolecules approaching, chemical bonds are formed between the active hydrophilic groups of the glutinous matrix and the reactive groups of glutinous macromolecules and hydroxyl groups on the surface of the filler [2, 5, 25].

The removal of moisture in the amount of 30% for a biocomposite material containing 100% wood flour leads to a decrease in the compressive strength to 28.7 MPa (Fig. 1), which is associated with a high degree of dehydration. This leads to a loss of fluidity of the glutinous binder during the formation of biocomposite samples at a molding temperature of 140°C. As a result, the number of chemical bonds between the components decreases, which is difficult to form under such conditions, which leads to a loss of strength. In the case of using 20% of crushed cereal stalks, the compressive strength increases to 103.5 MPa, which is associated with the ability to provide sufficient fluidity of the glutinous binder due to the ability of hydrophilic cereal stalks to accumulate water molecules. Grain stalk particles are 5–6 times larger in size (Fig. 4, *a*) compared to wood flour particles (Fig. 4, *b*), which complicates the removal of water from their surface layer during the treatment of the composition in a thermal field. This is confirmed by an increase in the compressive strength to 119.5 MPa in the case of an increase in the content of crushed stalks to 60% with a simultaneous decrease in the content of wood flour particles to 40%. In the case of an increase in the content of grain stalk particles to 80–100%, the tensile strength decreases by 20%, which is associated with a low content or absence of wood flour particles. Small wood flour particles provide a higher degree of compaction of the biocomposite material, since they fill the cavities in the contact points of large grain stalk particles. In the absence of small particles, an arched structure is formed (Fig. 8, *b*) from stem particles that have a small contact area and are characterized by a reduced number of chemical bonds.

The lowest impact strength (2.1 kJ/m²) is found in biocomposite materials containing 100% wood flour with a high moisture content in the composition (Fig. 9). As a result of removing moisture from the composition, this characteristic increases by 35%, which is associated with the formation of additional chemical bonds between the components of the system [5]. The impact strength of biocomposite materials whose compositions have lost 20% moisture is reduced by 18–20% compared to biocomposite materials whose compositions have lost 10% moisture. This is due to the increased rigidity of macromolecule chains as a result of the glutinous matrix dehydration process.

In the case of using a mixture of fillers consisting of 40% wood flour and 60% crushed stalks of cereals, the impact strength increases by 43–45% (Fig. 9) compared to biocomposite materials containing 100% wood flour. The increase in

impact strength is explained by the fact that the mechanical properties of polymer composites depend on the morphology and structure of the filler [11]. Processing such a composition in a thermal field enables the removal of moisture in the amount of 10%, resulting in an increase in impact strength [5] by 13–15% compared to biocomposites with an increased moisture content. Further removal of moisture from the composition in the amount of 20% leads to an increase in the resistance of biocomposite materials to the action of dynamic loads [2, 5]. Such materials have a high impact strength (10.2 kJ/m²) due to the formation of a dense structure of the biocomposite material with a compact arrangement of filler particles and a high number of chemical bonds between the active groups of the system components [11].

The devised technology for forming biocomposite materials with high mechanical properties could be used to produce packaging that is easily recycled without harming the environment. When using products based on biocomposite materials, it is necessary to control humidity and temperature conditions for the stability of the material as changes in humidity are decisive for their service life. The introduction of biocomposites makes it possible to reduce the plastic load on the planet and reduce the cost of production through the use of secondary raw materials.

The scientific novelty of our study relates to the first-ever technique for removing excess moisture from compositions based on glutinous binder. It provided the optimal degree of moisture at which the filler particles are compacted, the formation of chemical bonds between the components is intensified, and uniform removal of residues during heat treatment is achieved without the formation of voids and cavities.

Limitations on the technology of forming a biocomposite material include parameters for pressing and heat treatment of a composition with a fixed content of biopolymer binder and a certain combined content of fillers of different granulometric composition.

The disadvantage of the study is that our work did not determine the operational characteristics of the designed biocomposite material, which does not allow for an accurate prediction of its durability.

In the future, it is advisable to conduct a study on the operational properties of the designed biocomposite materials, in particular moisture, frost, and heat resistance, in order to determine their durability.

7. Conclusions

1. The maximum values of ultimate compressive strength (115–120 MPa) were obtained as a result of mixing fillers of different granulometric composition (40–80% of cereal stalk particles, the rest of the wood flour particles) provided that 30% of moisture was removed. Such results are attributed to the optimal moisture content in the composition, which ensures sufficient fluidity of the glutinous binder during the formation of the biocomposite material. Wood flour particles quickly lose moisture, which leads to low values of the ultimate compressive strength (28–30 MPa), compared to large cereal stalk particles that are able to accumulate moisture.

2. As a result of analyzing fracture fractograms, it was established that the removal of part of the moisture from the composition of the biocomposite material enables the formation of a biocomposite material structure with fewer

defects in the form of voids of size (80–100 μm). The removal of moisture at the stage of thermal treatment of the composition occurs due to the intensive removal of water molecules, which leads to the destruction of bonds between the components and the appearance of micropores 30–40 μm in size. Particles of cereal stalks are arranged chaotically in the glutinous matrix compared to the compact arrangement of wood flour particles. Therefore, under static loads, an uneven distribution of stresses occurs in local areas of the biocomposite material, which causes the appearance of cracks 150–200 μm in size.

3. The failure of biocomposite samples containing 100% wood flour occurs as a result of cracks appearing in the lower part of the cylindrical sample under the influence of elastic deformation, which indicates increased resistance to static loading. Biocomposite samples filled with cereal stalk particles plastically deform in the central part of the sample without the appearance of main cracks. This indicates increased strength of the biocomposite material, the composition of which contains 10% less moisture, compared to a similar sample, the composition of which has an increased moisture content. The failure of biocomposite samples containing 100% wood flour or 100% crushed cereal stalks and having lost 30% moisture in the composition occurs with the formation of several main cracks at an angle as a result of brittle fracture of the biocomposite.

4. Biocomposite materials containing 100% crushed cereal stalks have a 60–75% higher impact strength compared to biocomposites consisting of wood flour or a mixture of wood flour particles and crushed cereal stalks. Such materials are advisable to use for the manufacture of articles operated under dynamic loads. This is due to the resistance that cereal stalk fragments provide to crack propagation. In the case of article operation under static loads, it is advisable to use a mixture of fillers (wood flour and crushed cereal stalks) as a dense structure of the biocomposite material is formed with

a compact arrangement of particles of different particle sizes in the biopolymer matrix.

Conflicts of interest

The authors declare that they have no conflicts of interest in relation to the current study, including financial, personal, authorship, or any other, that could affect the study, as well as the results reported in this paper.

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Data availability

All data are available, either in numerical or graphical form, in the main text of the manuscript.

Use of artificial intelligence

The authors confirm that they did not use artificial intelligence technologies when creating the current work.

Authors' contributions

Vitalii Kashytskyi: Conceptualization, Methodology, Supervision; **Oksana Sadova:** Conceptualization, Methodology, Writing – original draft; **Valentyna Tkachuk:** Validation, Writing – review & editing; **Yevgeniy Chernota:** Investigation, Resources; **Nazarii Marchuk:** Investigation, Visualization.

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