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REINFORCING STARCH BIOPLASTICS WITH AGRICULTURAL WASTE

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Aim. The study was purposed to find alternative reinforcing fillers for the modification of starch-based bioplastics using agricultural waste.

Materials and Methods. The visual analysis method was used to compare the characteristics of materials with different types of fillers. The influence of various kinds of fillers on the mechanical properties and structure of starch-based bioplastics was evaluated. The study used corn starch according to the DSTU 3976-2000 standard and five different types of agricultural waste as fillers to modify biodegradable plastics. The method of manufacturing the bioplastics included preparing a 10% starch solution, mixing it with other fillers, heating the suspensions to 90 °C, and drying the resulting solutions at 60 °C, depending on the type of filler.

Results. The most successful options were those using technical cellulose fiber and sunflower seed husks compressed into granules. The obtained materials based on these fillers demonstrate better mechanical properties and better shape retention compared to starch-based materials without fillers. The optimal particle size was found to be in the range of 0.03-0.06 mm.

Conclusions. It can be concluded that agricultural vegetable waste has a high potential as an effective filler for starch-based bioplastics, which will significantly reduce the cost of biomaterials and expand the scope of their use, making them more accessible for a wide range of applications.

Key words: bioplastics, agricultural waste, reinforcing fillers, starch, properties.

The development of the bioplastics industry in Ukraine is an urgent issue, especially given the gradual abandonment of synthetic polymeric materials in the world. The lack of affordable foreign materials with reliable providers and national production stimulates further research in this area and the creation of local technologies [1].

The high cost is one of the main hurdles to the widespread use of bioplastics. Today, polylactic acid (PLA) and polyhydroxyalkanoates (PHA) are considered the most promising areas for the development of the biodegradable plastic market. However, the production of these polymers requires more sophisticated technologies than their synthetic counterparts, which significantly increases their cost [2].

Often, for reducing the cost and improving the mechanical properties of biodegradable materials, a certain amount of synthetic fillers is added to their composition. These are usually materials such as polyvinyl chloride or polyethylene. However, these components can negatively affect the biodegradability of polymers [3].

Therefore, the search for simplified technologies for the production of biodegradable materials using fillers of natural origin is relevant today [3].

Starch is one of the most readily available natural polysaccharides used to make biodegradable materials. An essential characteristic of starch-based bioplastics is their elasticity, which is provided by linear amylose, while amylopectin, which has a

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branched structure, controls tensile and elongation strength [4-5].

However, starch-based bioplastics are of limited use due to their insufficient strength. Therefore, modification using vegetable plasticizers, such as glycerin or sorbitol, as well as fillers that increase the mechanical strength and ductility of bioplastics, is necessary. In addition, the modification of hydrophilic OH-groups makes it resistant to moisture. Vegetable agricultural waste can be a promising alternative to synthetic fillers. Due to the high content of lignin and cellulose, these materials can be used as a reinforcing material to increase strength and stiffness for starch-based polymer metals [4].

The research aims to find reinforcing fillers for the modification of starch-based bioplastics using agricultural waste.

Materials and Methods

Materials

In the study of the process of obtaining biodegradable plastic based on starch, we used potato starch, which corresponds to DSTU 4286:2004 Potato starch. Technical conditions. Amended" [6].

To modify starch-based biodegradable plastics to improve their mechanical properties, the use of agricultural waste as a filler was investigated. For the study, five samples of fillers were selected:

– corn waste (dry stalks, leaves, cobs);

– waste peanut husks, crushed;

– sunflower husk pressed into pellets;

– technical fiber cellulose;

– crushed sunflower husk. The choice of specific types of plant waste for use as reinforcing fillers in starch plastic materials was based on their availability, physical properties, and economic advantages. The selected wastes include by-products of typical crops, so they are available in large quantities and have low costs. It is also important to note that obtaining fillers based on such materials can be an alternative method of their utilization and turn waste into a valuable resource [7]. All of these wastes have specific physical properties and chemical composition (high lignin and cellulose content), which can increase the strength and durability of bioplastics. The use of naturally occurring fillers helps to improve the biodegradation of polymeric material [7].

Corn waste (dry stalks, leaves, cobs). In terms of chemical composition, this type of raw material contains about 43% cellulose, 18% lignin, 26% pentosans, 4% inorganic substances, and 15–18% moisture [7, 8].

Peanut husks contain about 37–45% cellulose, 28-35% lignin, and 18–30% hemicellulose. Due to this composition, it is considered a promising raw material for biotechnological applications, in particular in the production of bioplastics, biofuels, and fermented products. To improve the fusion of this type of filler with starch-based biodegradable plastics, it is crushed.

Sunflower husk pellets (produced by Harveles OIL) are compressed cylindrical pellets 5 cm long and about 8 mm in diameter. The moisture content of the pellets does not exceed 8.6% [9].

Crushed sunflower seed husks. The chemical composition of the husk may vary depending on the sunflower variety, with high-oil species having a higher fat and ash content but lower fiber content. Sunflower husk composition: lipid content for different varieties ranges from 0.99 to 3.78%, fiber — 49–67%, of which cellulose — 31–42%, and 14 pentosans — 23–28%. The amount of lignin in the husk is 24–30%, and the proportion of inorganic components is 1.37–2.98% [10].

Fibrous technical pulp was also used as a filler. Cellulose has a fibrous structure and is insoluble in water. It was obtained from vegetable, mechanically processed raw materials by sulfite and alkaline cooking.

Methods

Methods of obtaining biodegradable materials based on starch

The technological properties of starch materials are still inferior to polyethylene and polypropylene, which they could replace. Materials made from starch are pretty fragile and not resistant to temperature or moistureю. However, they can provide the necessary flexibility to other biopolymers. Starch-based polymers can also be used as a separate type of material, but they must be reinforced with fillers to provide mechanical strength and stability [4].

The methodology for producing starch-based bioplastics consists of several steps. First, a 10% starch solution is prepared in a 100 cm^3 flask. 10 g of starch and 60 cm^3 of distilled cold water are mixed, stirred for 15 minutes to ensure uniform distribution of starch in the dispersion medium. Following starch dispersion, 5 cm^3 of glycerol is added, and the volume is brought up to 100 cm^3 with distilled water. The resulting mixture is subsequently stirred for an additional 5 minutes [5].

The 100 cm^3 of starch was divided into five portions for mixing with the appropriate fillers. To 20 cm^3 of the prepared 10% suspension, 1 g of crushed fillers of various types was added [5].

Next, the resulting suspensions were heated to 90 °C with constant stirring for 25 minutes until a homogeneous solution with slight opalescence was obtained; the filler particles should be evenly distributed in the solution. [5].

The resulting solution was evenly distributed in a 5 mm thick layer on a flat surface and placed in a drying oven preheated to 60 °C. Drying was carried out with different durations for different types of filling: 3.5–6 h. [5]

To evaluate the biodegradation potential of polymeric materials, a vermicomposting method utilizing household organic waste and the earthworm species *Eisenia fetida* was employed. The vermicomposting conditions were maintained within a temperature range of 15–25 °C, a moisture content of 70–80%, and a pH between 7.0 and 7.6.

Starch-based polymeric materials were subjected to the vermicomposting process in order to determine their susceptibility to biodegradation. The results demonstrated a high degree of biodegradability, with complete decomposition of the materials observed within 60 days. The selected filler types demonstrated a high degree of compostability under the established vermicomposting conditions..

Results and Discussion

A polymeric material based on starch was obtained. A visual assessment of the material was carried out. According to which we can say that this material is hard, transparent, very elastic, not viscous, brittle, and easily crushed. The drying time of the material at a temperature of 60 °C is one hour. The main disadvantage observed in comparison with synthetic analogues is low strength and poor shape retention.

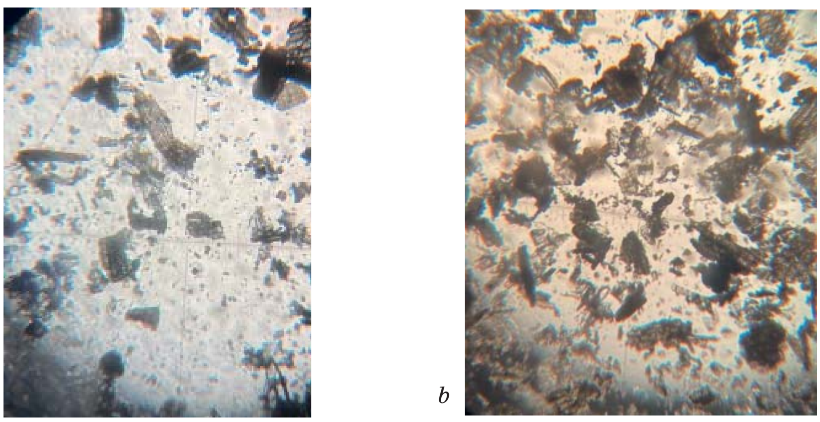
To improve the mechanical properties of the resulting material, crushed materials of plant origin were used. The following results were obtained by grinding different types of fillers.

The first type of raw material, based on corn stalks, leaves, and ears, is well crushed, and the resulting mixture is homogeneous. To determine the size of the ground particles, they were microscoped using a ULAB XSP-137 biological microscope; the results are shown in Fig. 1. The average particle size is 0.03–0.025 mm.

Another type of raw material — crushed peanut husks — is characterized by much better properties during mechanical processing. The degree of grinding is maximized, the resulting mixture is homogeneous, and the particles are easily ground to a powder. The average particle size is 0.05–0.08 mm, and the presence of larger fragments is not observed. To confirm the homogeneity and degree of grinding, the samples were microscoped using a ULAB XSP-137 biological microscope. The results of the study are shown in Fig. 2.

The raw material is based on sunflower husks, which are easily crushed, and a powder with a uniform distribution of particles was obtained.

To determine the size of the crushed particles, they were microscoped using a ULAB XSP-137 biological microscope. The results are shown in Figure 3. The average size of the crushed particles is 0.04–0.07 mm.



a *b*

Fig. 1. **Microscopy of crushed corn waste particles (ULAB XSP-137), ×10 objective and ×10 eyepiece, division price is 0.01 mm**

Sunflower husk pellets with a diameter of 8 mm and a length of 2–5 cm were crushed to improve the fusion of this type of filling with starch-based biodegradable plastic. As a result of grinding, a heterogeneous mixture was obtained. The smallest particles have a size of 0.03–0.05 mm; there are also fibers with a width of 0.002 mm, and the largest particles are within 2–5 mm. To determine the size of the crushed particles, they were microscoped using a ULAB XSP-137 biological microscope; the results are shown in Fig. 4.

The technical fiber pulp was not subjected to additional grinding, as the fiber size in its original form is sufficient for mixing with the polymer material. The fiber thickness was approximately 0.01 mm so that the length could vary significantly. To determine the size of the cellulose fibers, microscopy was performed using a ULAB XSP-137 biological microscope; the results are shown in Fig. 5.

Images of the materials obtained by mixing starch-containing polymer with various types of fillers are shown in Fig. 6.

A visual analysis of the differences in the characteristics of the materials was carried out, and the method of measuring Shore hardness (Shore hardness) was used using a durometer, which measures the ability of a material to resist the penetration of a steel pin into its surface. The following scales were used scale A — to measure the hardness of elastic materials such as rubber or soft plastics. Based on the data obtained, a conclusion was made about the uniformity of particle distribution in the polymer, as well as its hardness, strength, and ability to retain its shape.

Method in accordance with ISO 868-85 Plastics and ebonite. Determination of indentation hardness by durometer (Shore hardness).

Table 1 shows the measured hardness values for each of the samples.

The characteristics of the obtained samples of polymeric materials with different types of fillers are shown in Table 2.

A comparative analysis of starch-based polymeric materials with various plant-based additives revealed significant differences in their structural and mechanical properties, including surface texture, uniformity, and Shore A hardness.

The control sample, consisting of pure starch-based bioplastics without fillers, showed the lowest hardness value (13.87 A). Despite its high elasticity and smooth appearance, its mechanical stability and ability to retain its shape were significantly limited. This is in line with previously reported data on the inherent softness and flexibility of starch-based polymers in the absence of reinforcing agents.

Among the modified composites, the inclusion of sunflower seed husks compressed into pellets resulted in the highest hardness (47.00 A), indicating excellent mechanical strength. However, visual analysis revealed a coarse and granular structure, which, while effective in increasing stiffness, compromises material surface uniformity and elasticity. This filler has proven to be optimal in applications that require structural stability over flexibility.

The sample filled with crushed peanut husks also showed a significant improvement

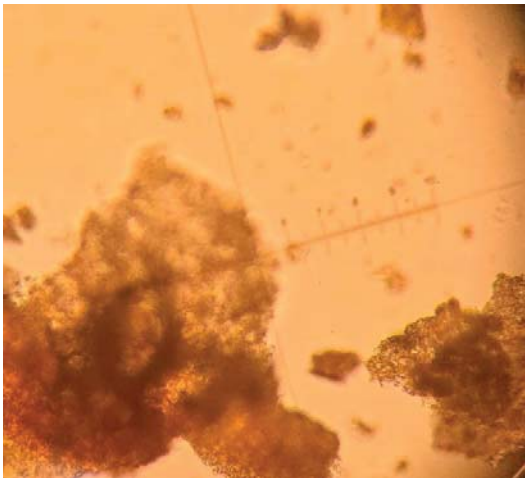


Fig. 2. **Microscopy of crushed peanut husks (ULAB XSP-137), ×10 objective and 10x micrometer eyepiece, division price is 0.01 mm**

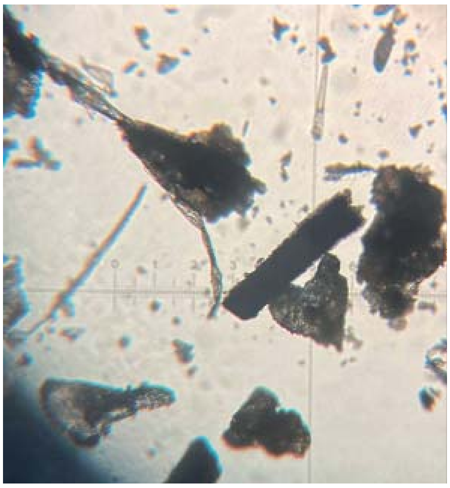


Fig. 3. **Microscopy of crushed sunflower seed husk particles (ULAB XSP-137), ×10 objective and ×10 micrometer eyepiece, division price is 0.01 mm**

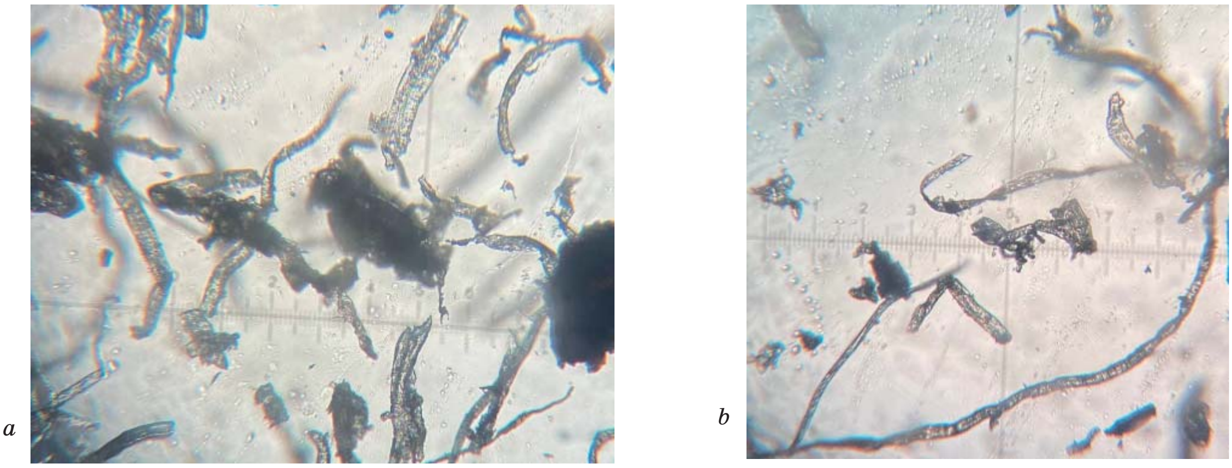


Fig. 4. **Microscopy of particles of crushed sunflower husk pellets (ULAB XSP-137), ×10 objective and ×10 micrometer eyepiece, division price is 0.01 mm**

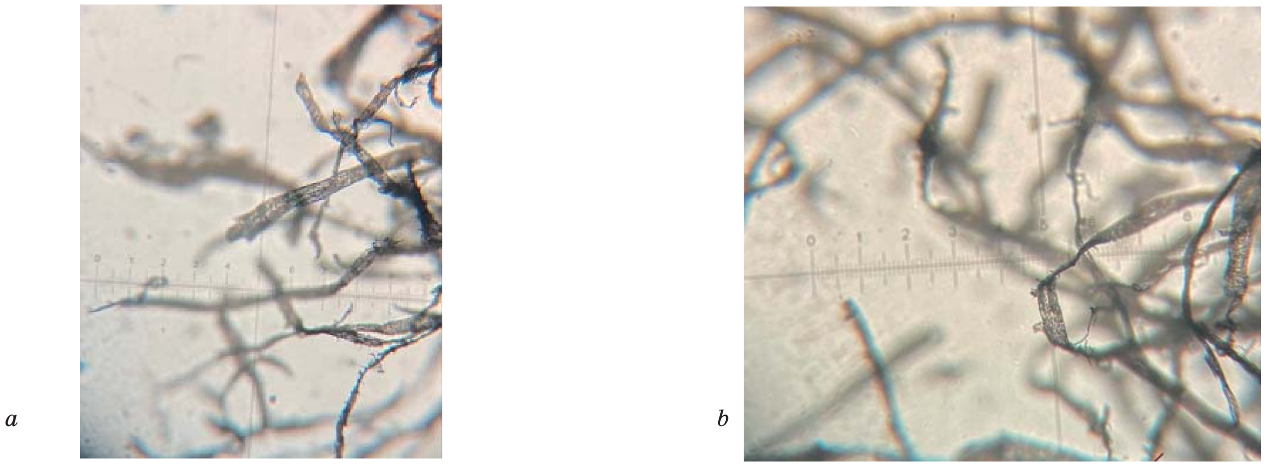


Fig. 5. **Fiber microscopy (ULAB XSP-137), 10× objective and 10× eyepiece, division price is 0.01 mm**

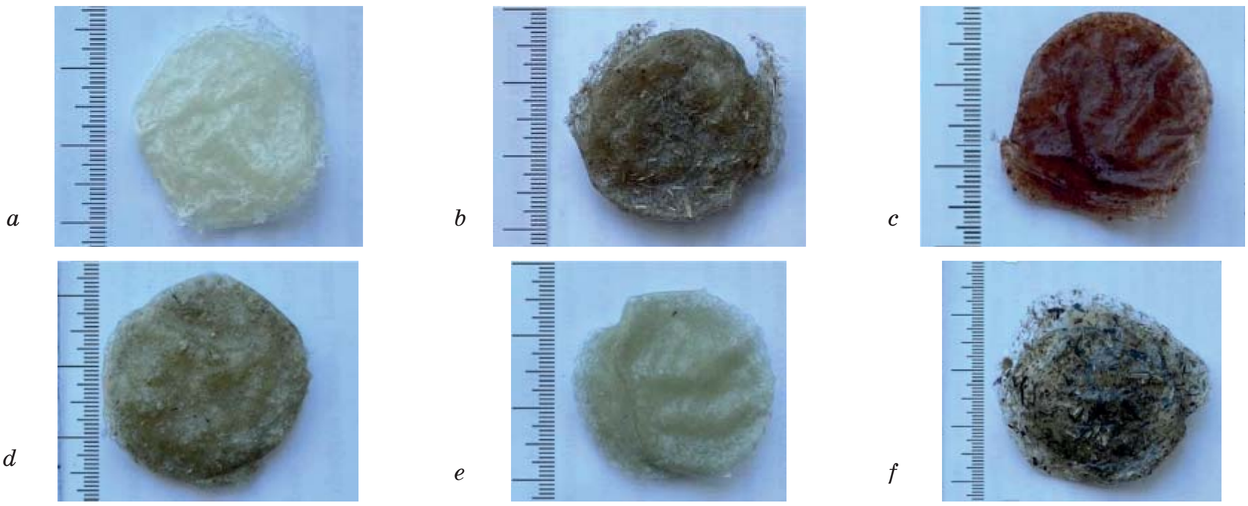


Fig. 6. **The obtained polymeric materials reinforced with different types of fillers based on crushed agricultural waste**

a — starch-based bioplastic without fillers; *b* — corn waste (dry stems, leaves, cobs); *c* — crushed peanut husks; *d* — sunflower husks compressed into granules; *e* — technical cellulose fiber; *f* — crushed sunflower husks

Table 1

Hardness values for each of the samples

Type of filler	**Hardness indices in three repetitions. A**	**Average hardness value**
starch-based bioplastic without fillers	16.1	13.87
	10	
	15.5	
corn waste (dry stems. leaves. cobs)	20.5	28.5
	24.5	
	40.5	
crushed peanut husks	35	32.83
	36	
	27.5	
sunflower husks compressed into granules	49.5	47.00
	46.5	
	45	
technical cellulose fiber	15.5	16.5
	18.5	
	15.5	
crushed sunflower husks	39.5	39.5
	42.5	
	36.5	

in hardness (32.83 A) compared to the control. Despite the somewhat uneven distribution of particles, the material retained a relatively balanced structure with acceptable shape retention and reduced brittleness. This indicates that finely ground peanut husks can serve as an effective reinforcing agent, especially in semi-solid biopolymer products.

The corn waste (dry stalks, leaves, and cobs) had a moderate hardness level (28.5 A), a more visually uniform structure, and a smoother surface texture. This filler resulted in a material that successfully combines stiffness and light flexibility, making it suitable for flexible packaging or biodegradable sheets.

In contrast, the samples filled with chopped sunflower husk (39.5 A) and technical cellulose fiber (16.5 A) showed mixed results. While the crushed sunflower husk contributed to the hardness increase, its poor distribution due to the coarse particle size resulted in localized brittleness and poor mold stability. On the other hand, industrial cellulose fiber improves material homogeneity and reduces brittleness, but does not significantly increase mechanical strength.

Conclusions

In the context of the ever-increasing awareness of environmental issues and changes in the global plastic materials market, the development of bioplastics production is becoming an urgent task, especially for countries like Ukraine. The research conducted in this paper aims to find alternative reinforcing fillers for the modification of starch-based bioplastics using agricultural waste.

The study results showed that the use of different fillers significantly affects the mechanical properties of starch bioplastics. The best results in terms of stiffness (47.00 A) and mechanical strength were obtained with sunflower husk granules. However, this filler has a coarse structure, which reduces the elasticity and homogeneity of the material, making it more suitable for applications where mold stability is essential.

Other materials, such as crushed peanut husks (32.83 A) and corn stover (28.5 A), showed moderate results, combining stiffness and elasticity. The crushed peanut husks provided good uniformity and reduced brittleness, while the corn stover was a material that combined flexibility and sufficient stiffness for biodegradable films and packaging.

Cellulose, although it does not provide a significant increase in mechanical strength (16.5 A), improves material uniformity and reduces brittleness. It is a good choice for products where a stable structure is important without high stiffness requirements.

Prospects for further research are to refine technological processes to improve the distribution of particles in the material and to study resistance to temperature, hydrolysis, and biodegradation. This will allow us to expand the use of bioplastics and ensure their stability in various conditions, in particular for the production of durable and environmentally friendly products. Based on our analysis, starch-based bioplastics reinforced with agricultural plant waste show promising potential in the following applications.

Short-term packaging, such as disposable food trays. The material demonstrates sufficient mechanical strength for food contact and is fully compostable, including through vermicomposting.

Table 2

Description of the properties of the obtained starch-based polymeric materials using different types of fillers

Type of filler	Properties of the resulting material	Time. h
Starch-based bioplastics without fillers	Semi-transparent material with a glossy surface, without visible inclusions. Demonstrates good elasticity, but has the lowest hardness among all samples (13.87 A), indicating low shape retention and mechanical instability under deformation.	3.5
Corn waste (dry stalks, leaves, ears);	Translucent yellowish sample with a homogeneous texture. The high degree of grinding ensured a uniform distribution of particles in the polymer. Still, there are particles of different shapes and sizes, which can cause insufficient resistance to deformation. It has a hardness of 28.5 A, which allows it to retain its shape better than bioplastics without fillers, while maintaining partial flexibility.	5.5
crushed peanut husks	The material is dark in color with noticeable particle inclusions. The sample has a hardness of 32.83 A, which indicates sufficient strength and shape retention, The husk particles are evenly distributed, which makes the material somewhat brittle at the edges, but generally holds its shape well.	3.5
Sunflower seed husks are pressed into pellets;	One of the most complex samples (47.00 A). The material is dark, with a rough texture and granular inclusions. The filler particles are embedded in the polymer matrix quite densely, but the insufficient degree of grinding reduces the overall resistance to damage. The material is inelastic but retains its shape very well.	5.5
Technical fiber pulp;	Translucent sample with a fibrous structure. The fibers are well distributed, which reduces brittleness. The material has medium hardness (16.5 A) and is stable in shape. Due to the good distribution of structural particles in the material, it has a good ability to retain its shape. It is suitable for the formation of rigid films that maintain their geometry under load.	5
Crushed sunflower seed husks;	A material with a high level of hardness (39.5 A) but an uneven distribution of particles. The surface is rough, with noticeable inclusions of husks. As a result, the sample does not retain its shape well under load and is prone to deformation. Low elasticity and increased brittleness reduce functionality in flexible applications.	4

Agricultural use, including biodegradable seedling pots and mulching films. After fulfilling their function, these materials can be left in the soil, where they naturally decompose without leaving harmful residues.

Authors' contribution

Kozar M.Y. — data collection and original draft preparation; Korneliuk O.A. — investigation, formal analysis. The authors have read and agreed to the published version of the paper.

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Conflict of interest

The authors declare no conflict of interest.

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АРМУВАННЯ БІОПЛАСТИКІВ НА ОСНОВІ КРОХМАЛЮ СІЛЬСЬКОГОСПОДАРСЬКИМИ ВІДХОДАМИ

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Мета. Пошук альтернативних армуючих наповнювачів для модифікації біопластиків на основі крохмалю з використанням відходів сільського господарства.

Матеріали й методи. Для порівняння характеристик матеріалів з різними типами сільськогосподарських відходів як наповнювачів використано метод візуальної оцінки та Метод вимірювання твердості за Шором. Оцінено вплив різних типів наповнювачів на механічні властивості та структуру біопластиків на основі крохмалю.

У дослідженні використовували кукурудзяний крохмаль за стандартом DSTU 3976-2000 та п'ять різних типів сільськогосподарських відходів як наповнювачі для модифікації біорозкладних пластиків. Метод виготовлення біопластиків включав підготовку 10% розчину крохмалю, змішування його з різними наповнювачами, підігрівання суспензій до 90 °C та сушіння отриманих розчинів при 60 °C залежно від типу наповнювача.

Результати. Найбільш вдалими виявилися варіанти з використанням технічного волокна целюлози та спресованого в гранули лушпиння соняшникового насіння. Отримані матеріали на основі цих наповнювачів демонструють кращі механічні властивості та краще збереження форми порівняно з матеріалами на основі крохмалю без наповнювачів. Оптимальний розмір частинок виявився в діапазоні 0,03–0,06 мм.

Висновок. Рослинні сільськогосподарські відходи є ефективними для біопластиків на основі крохмалю, що дозволить значно знизити собівартість біоматеріалів та розширити сферу їх використання, зробивши їх більш доступними для широкого спектру застосувань.

Ключові слова: біопластик, сільськогосподарські відходи, армуючі наповнювачі, крохмаль, властивості.