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Obtaining compostable composites from secondary raw materials of crop production

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Abstract. Proceeding from the goals of sustainable development of the United Nations and the active introduction of the principles of the circular economy's concept, the possibility of using the secondary raw and material resources in the production processes is relevant. At the UN Environment Assembly in 2022 representatives of 175 countries approved a resolution and called for a joint effort of states to reduce plastic pollution. Biodegradable plastics are a well-known alternative to traditional plastics with a long natural decomposition period. Among biodegradable materials, compostable materials are especially interesting due to the possibility of absolute degradation to biomass, CO_2 and water. The problem of obtaining compostable materials is related to the possibility of using secondary material resources and achieving certain quality parameters of the final product. In this research, the criteria and technological solutions for obtaining compostable materials for single use are reviewed. The requirements to the raw plant material and the final product were analyzed, various ways of obtaining such materials and raw material processing methods were investigated. As a result of the work, the ratios of the mixture components were selected, the particle sizes of the plant component were determined, the use of a hot press machine was validated and the technological parameters for obtaining the finished form of the material were determined. The obtained results of the study are planned to be used for the development of production technology of compostable bio-composites and its further scaling for their mass production of bio-composite materials for consumer and industrial needs.

Key words: biodegradable plastics, compostable composites, reduce plastic pollution, secondary raw resources, plant component.

INTRODUCTION

Most petroleum-based plastics are highly resistant to degradation in nature, being able to persist for hundreds of years in the environment, and have a negative impact on the environment (Abraham et al., 2021). On 2 March 2022, the United Nations Environment

Assembly adopted Resolution 5/14 'End plastic pollution: Towards an internationally legally binding instrument'. The main problems highlighted in the resolution are the high and rapidly increasing levels of plastic pollution in the environment and in the world's oceans, and the proliferation of microplastics. These problems and their consequences are transboundary in nature and need to be addressed through a life-cycle approach that takes into account national circumstances and capacities. In this context, a shift towards sustainable production of biobased polymers is required.

Biobased plastics and biodegradable polymers are now the main substitutes for traditional plastics, as evidenced by the growing market, which now represents over 1% of the total plastics produced at 368 M tonnes annually (Undas et al., 2022). Biodegradable polymers can be recycled using various technologies, for example aerobic composting, anaerobic decomposition, agricultural mulching, and solubilisation, that lead to reduced waste generation and soil contamination (Razza et al., 2020).

The main challenge in obtaining new biodegradable polymers is to find alternative raw materials that meet the criteria of environmental safety, recyclability, as well as economic efficiency. Non-food crops, agricultural residues, forest residues, microbial polymers, polymerised monomers, and agri-food waste are being considered most actively.

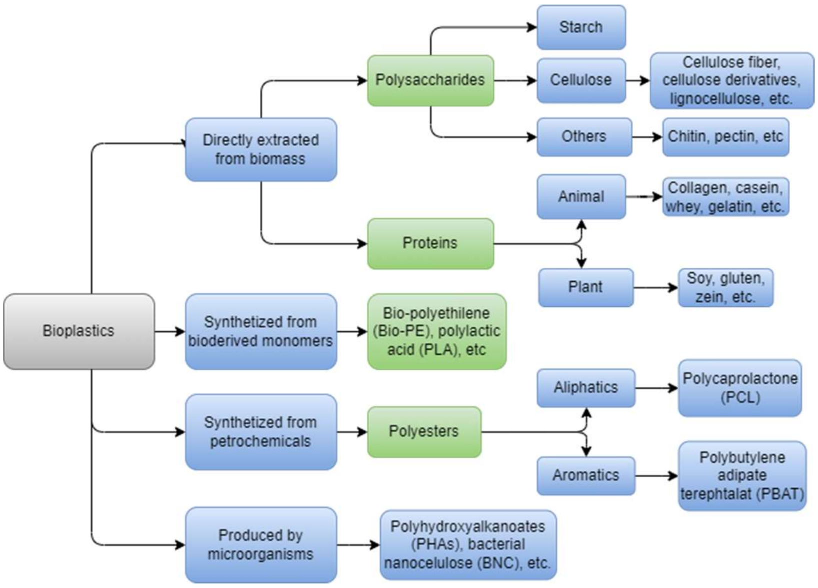


Figure 1. Classification of bioplastics by origin.

One type of bioplastics are compostable composites, which are polymerised polysaccharides with the possibility of incorporating reinforcing materials to impart strength properties. In this regard, numerous studies have been conducted to find materials and technologies for their production. In particular, ways of processing agro-industrial wastes and by-products for bioplastics synthesis (Khajuria, 2017; Sadhukhan et al., 2018; Jõgi & Bhat, 2020; Mirpoor et al., 2021), direct biopolymers, and extraction of active compounds from agricultural residues (Fig. 1) (Julieta et al., 2016; El Barnossi et al., 2020; Shachi et al., 2021) were considered. Rice husks, cassava roots, sugar cane, corn husks, orange pomace, malt pomace, sesame seed cake and other materials were suggested as reinforcing additives for composite materials (Teixeira et al., 2011; Phiriyawirut & Maniaw, 2012; Vercelheze et al., 2012; Bodirlau et al., 2013; Versino et al., 2015; Machado et al., 2017; Gunarathne et al., 2018; Ferreira et al., 2019).

The present work is devoted to the development of compostable composites using secondary raw materials from crop production. The formulation of the created materials and the technology of their production are proposed.

Among biopolymers, starch is one of the most promising renewable materials because of its versatility, inexpensive price, abundance, and biodegradability. Natural starch can be converted into thermoplastic starch by heating and breaking down starch granules in the presence of a plasticizer (usually water and a polyol, such as glycerol or sorbitol). Under these conditions, the starch granules break down, plasticise and melt to form a material whose behaviour is similar to that of thermoplastics (Dufresne et al., 2000; Avérous & Halley, 2009; Hietala et al., 2013). Therefore, starch has been used to produce biodegradable plastics, which are mainly used in films for food packaging, household bags, and flushable materials for hygiene products (Avérous & Boquillon, 2004). However, to obtain starch biocomposites (SBs) with better properties, several limitations of this material, in particular its poor mechanical properties, need to be overcome. This issue can be solved using several approaches, such as incorporating plasticizers, blending starch with polymers, chemically modifying starch, and adding fibres and nanoparticles to improve mechanical properties (Schlemmer et al., 2010; Ashogbon & Akintayo, 2014; Ibrahim et al., 2014; Lomeli Ramirez et al., 2014; Cano et al., 2015; Ortega-Toro et al., 2015).

Obtaining a composite involves the addition of reinforcing material to enhance the strength characteristics of a material. This is a common method to improve the properties of starch composites. Various types of natural fibres for starch-based compostable composites have been reported in the literature, including kenaf fibre (Zainuddin et al., 2013), oil palm fibre (Mahmood et al., 2016), loofah fibre (Kaewtatip & Thongmee, 2012), jute and kapok fibre (Prachayawarakorn et al., 2013), and cotton (Prachayawarakorn et al., 2011), as well as husks, hulls and peels (Shabarin et al., 2021).

In our work, we considered the use of rice husks and barley bran as fillers for the biocomposite. Rice husk is a natural biopolymer with 75–90% organic matter content, including cellulose and lignin, and the remaining mineral components such as silica (Kargarzadeh et al., 2017). The presence of silica, in addition to cellulose, makes rice husks a particularly relevant raw material for biocomposites based on their reinforcing strength characteristics. The prevalence, physical and technological properties make rice husks a suitable raw material for the production of biocomposite compostable materials with high physical and mechanical properties.

Barley husk is a waste product of one of the seven most common cereal crops. Global barley production in 2019 was 159 million tonnes. (Kärkönen et al., 2022). The lignin-cellulose waste presents about 66% of the total production and has a low added value. The literature analysis has shown that the contained cellulose of the fiber is the main component responsible for the mechanical strength of fiber-reinforced composite films (Mittal et al., 2016). Our work considers the use of barley husk as a filler to produce a compostable composite material.

Polylactide (PLA), which is derived from renewable sources with high carbohydrate content, such as sugarcane (Okada, 2002), is a thermoplastic biocompostable polymer. Due to its biocompatibility, it is also used in biomedicine. Polylactide has hydrophobic properties, good transparency and high stiffness, which allows its use as a laminating coating. Due to high demand for biodegradable materials, products using PLA, such as film, food packaging, textiles, bottles and utensils, have been produced in recent years in many countries (Auras et al., 2004; Farrington et al., 2005; Lee et al., 2005). The high melting point (around 175 °C) provides thermal stability of the resulting products, and the proven performance of PLA biodegradable to CO_2, water, and biomass under controlled aerobic industrial composting conditions in less than 90 days explains the interest in this type of composite (Tokiwa & Calabia, 2006; Teixeira et al., 2011).

MATERIALS AND METHODS

Rice and barley husks ground to a particle size not exceeding 0.20–0.25 mm were used as secondary raw materials from crop production to produce compostable composites. Polylactide (PLA) was used to impart barrier properties (thermal resistance and hydrophobicity) to the composite material.

To obtain the mixture, potato starch according to (GOST R 53876-2010) was used, which was subjected to gelatinization in water in the ratio of 26% using direct heating in a water bath, and 37.14% using a microwave oven. The obtained mixtures were shaped into a flat plate of 1 mm thickness by pressure roller. The drying process of the plate was carried out by convection drying in a thermostat. The mixture was shaped into a 1 mm thick flat plate by hand rolling with a pressure roller. The drying process was carried out by convection drying in a thermostat.

Extrusion was used to heat treat the composite, remove excess moisture and shape it into a flat sheet. This method has an advantage in terms of performance and efficiency of composites processing using plant-based raw materials (Debiagi et al., 2010; Kesari et el., 2022).

Extrusion was performed at a temperature of 50–67 °C, 770–800 RPM, at a pressure of 1-2 MPa. The pre-prepared layer of biodegradable material was dried and ground to a fine particle size of 2–3 mm. To dry, increase density and obtain a uniform material thickness, hot pressing was applied using a Romanoff vulcanising press with a plate size of 20x13 cm, 700 W, and a maximum heating temperature of 288 °C. The vulcanizer had an exposure temperature of 120 °C, an exposure time of 3 min, a mould for pressing of 10×8 cm and thickness of 1 mm.

Studies (Aadil et al., 2016) and (Kwong et al., 2014) were analyzed to develop a methodology for determining the ability to preserve the shape in contact with water and the time of obtaining composite materials. In our experiment, before examining the samples, the moisture content in the samples was determined using an OHAUS MB-25 humidity analyzer. Samples weighing 0.4 g were placed in 150 mL of water after rolling with a pressure roller with exposure in a thermostat or after extrusion. To determine the time of destruction, the samples were completely immersed in water at a temperature of 20 °C and 80 °C for 2 h without stirring. The time of the beginning of dissolution and complete dissolution of the samples was measured, and the degree of dissolution was assessed by visual observation. After a comparative analysis, the mechanical strength of the materials under study was determined.

In this study, the tensile strength and elongation values of the material were determined according to (ISO 527-2:2012).

In (Castanho et al., 2021) starch and glycerine were used as binders and rice hulls as fillers to make a composite biodegradable mixture. In (Wilpiszewska & Antosik, 2022) citric acid was used to improve the gelatinization of starch. Based on the data from these works, we developed the following formulation shown in Table 1.

Table 1. Recipe for compostable biocomposite material produced using water bath

Component	Quantity	
	g	%
Filler	25	25.91
Glycerine	8	8.29
Water	50	51.81
Citric acid	0.5	0.52
Starch	13	13.47
Total:	96.5	100

Having analyzed (Spada et al., 2020), a proprietary methodology was developed to produce a compostable composite material.

A water bath was performed with constant stirring at a temperature of 90 °C for 15 min until the mixture became viscous. To prepare the gelatinized mixture, potato starch (13.47%) and citric acid (0.52%) were added to glycerol taken in an amount of 8.29% of the total mass of the composition, poured with water (51.81%) and mixed. Immediately after removing the starch hydromodule from the water bath, the filler (25.91%) was added, homogenized, and shaped. The resulting samples were then placed in a thermostat at 65 °C for 3 h. The recipe of the obtained material is presented in Table 2.

Table 2. Recipe ratios of the composite components obtained using water bath (R.H. – rice husk, B.H. – barley husk)

No.	Starch, %	Water, %	Glycerine, %	Citric acid, %	Filler RH, %	Filler BH, %
1	13.47	51.81	8.29	0.52	25.91	–
2	13.47	51.81	8.29	0.52	19.43	6.47
3	13.47	51.81	8.29	0.52	12.955	12.955
4	13.47	51.81	8.29	0.52	6.47	19.43
5	13.7	51.81	8.29	0.52	–	25.91
6	14.21	49.18	8.74	0.55	27.32	–
7	15.03	46.24	9.25	0.58	28.90	–
8	15.95	42.96	9.81	0.61	30.67	–

To determine the tensile strength and relative elongation of the obtained samples, tests were carried out in accordance with the (ISO 527-2:2012) method.

As a result of experiments and research analysis (Castanho et al., 2021), a methodology was developed, and a faster and more energy efficient method of gelatinization of the formulation mixture using a microwave oven was tested. The water content was reduced to optimum for gelatinization due to the reduced moisture content during evaporation in the microwave oven. During the laboratory studies it was also decided not to use citric acid and glycerine in the formulation for the mixture to achieve the expected physical and mechanical properties of the final material and to reduce the production costs.

Gelatinisation using a microwave oven was carried out at a power of 800 W. A 6.9% rice husk weight was added to the tank and a 17.8% starch weight was added. Then, water with a temperature of 96 ± 2 °C was added and the contents were stirred to a homogeneous mixture. The resulting mixture was placed in a microwave oven for 30 s, stirring every 10 s. After the mixture was gelatinized, the remaining 27.4% rice husk was added and stirred until the mixture was homogenized again. The resulting material was manually shaped into a globular shape and rolled with a pressure roller to form a flat plate 1 mm thick. Table 3 shows the developed formulation of the compostable material obtained using a microwave oven.

Table 3. Recipe for compostable biocomposite material produced using a microwave oven

Component	Quantity	
	g	%
Filler	25	34.25
Water	35	47.95
Starch	13	17.8
Total:	73	100

To achieve the expected physical and mechanical properties required for the final compostable product, such as tableware, it is necessary to coat the material with a thin layer of polymerised lactic acid (polylactide, PLA) in the form of a laminating coating. The estimated amount of polylactide is 30% of the material mass used to manufacture 1 workpiece.

Extrusion was carried out at a temperature of 50–67 °C, 770–800 RPM, at a pressure of 1–2 MPa. The pre-prepared layer of biodegradable material was dried and ground to fine particles, 2–3 mm in size.

As an alternative to the manual rolling method followed by convection drying, a hot pressing method was used to dry, increase the density, and obtain a uniform thickness of the material. The pre-prepared compostable material was hand-rolled to a thickness of 1 mm and subjected to pressing at 120 °C for 3 min using a Romanoff press vulcaniser. The size of the substrate (pressing mould) was 10x8 cm. A sample of 1 mm thick PLA lamination plate was also prepared at 200 °C for 30 s. The resulting PLA plate was fused to the compostable material for 1 min at 200 °C to form a strong composition (Fig. 4).

RESULTS AND DISCUSSION

The results of tests carried out to determine the tensile strength and relative elongation of the samples obtained are shown in Table 4. The results for the materials from which it was not possible to prepare samples in accordance with the methodology due to their fracture are not shown.

In obtaining the compostable material, it was found that the samples in which barley husk was used as the main filler were more hygroscopic than those using rice husks.

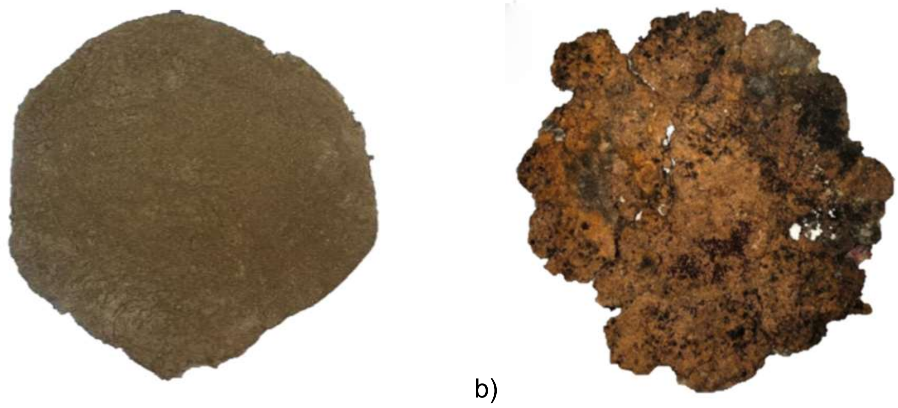
The material using barley husk did not mix up, which left its structure heterogeneous, and the material did not homogenize with the mixture components and remained on the substrate when separated. It was also found that, barley husk materials had increased

Table 4. Comparative characteristics and values of tensile strength and relative elongation of the material

No. [1]	Material characteristics	Strength, MPa	Elongation, %
1	Medium moisture, brittle andelastic, sticking to paper	–[2]	–[2]
2	Medium moisture, brittle andelasticity	0.49	0.60
3		0.32	0.47
4		0.22	0.35
5	High moisture and brittle, lowelasticity	–[2]	–[2]
6	Low humidity and fragility, highelasticity	0.53	0.60
7		0.57	0.63
8		0.62	0.65

[1] Sample numbers correspond to the composition given in Table 2; [2] '–' means not possible to prepare the sample for testing due to sample failure.

hygroscopicity, as shown in Fig. 2, b. Thus, the use of rice husk as the main filler in the production of compostable material (Fig. 2, a) was found to be the most rational by experiment. Fig. 2 shows the samples obtained according to the formulations presented in Table 2.



a) b)

Figure 2. Compostable material: a) sample formulation No. 8; b) sample formulation No. 5.

As a result of the series of experiments, an optimized mixture formulation was made to produce compostable materials (Table 3). After gelatinization of the mixture, the resulting material was rolled manually with a pressure roller to a plate 1 mm thick. The rolled out sheet of material was then shaped by hand into final products - plates with high walls. The product was then placed in a convection drying oven at 65 °C until completely dry (Fig. 3).

Figure 3. Plate from sample recipe No. 8.

When tests were carried out to select a method for producing compostable material using an extruder, the formulation mixture in the machine started to char when the temperature rose above 68 °C. The friction of the extruder screw exceeded the acceptable values, resulting in drastic reduction in speed and an increase in the load on the electric motor during material production. The tests showed that it was not practical to use the extruder for moulding due to the characteristics of the mixture.

After extruding the material, a series of experiments were performed to compare the properties of the samples obtained, with those produced by hand-rolling. The results of the experiments are demonstrated in Table 5.

The compostable composite obtained by hand rolling pressure roller did not lose its shape when kept in water at a temperature of 20 °C for more than 2 h, unlike the extruded composite, which began to lose shape after 10 min, and completely collapsed after 40 min from the beginning of the research. The composite, twisted by hand rolling pressure roller, began to change its appearance after 30 min of exposure in water at a temperature of 80 °C and completely lost its shape after 2 h. Under similar conditions, the extruded composite began to collapse almost immediately after immersion in water, and completely lost its shape after 15 min. The strength of the extrusion composite is significantly lower than that of the rolled composite, which is resistant to mechanical stresses.

Table 5. Comparative characteristics of materials obtained by different methods

Comparison index	Rolling by pressure roller	Extrusion
Moisture	4.96%	2.9%
Changing the shape in water	Didn't change the shape	Begins to lose the shape after 10 min, and completely collapsed after 40 min
Changing the shape in hot water	Begins to change the appearance after 30 min, and completely lost the shape after 2 h	Completely lost the shape after 15 min
Mechanical strength	Resistant to mechanical impact	The material is brittle
Strength, MPa	0.62	0.25
Elongation, %	0.65	0.29

Therefore, processing by extrusion has led to a significant deterioration of the material properties, which can have a negative effect in future. Thus, extrusion is inappropriate for moulding the composite of the mixture under study, as opposed to the thermo-pressing method, which produces a uniformly dried at 120 °C for 3 min thin layer of compostable material.

To improve the thermal and hydrophobic properties of the resulting compostable materials, the need for laminating the blanks was identified in the process of developing the composting technology. A 1 mm thick layer of PLA was applied to a compostable blank (formulation sample No. 8) (Fig. 4). To prepare for lamination, the PLA was heated on a thermal press to 200 °C for 1 min. To obtain a laminated sample of the compostable material, the blank was joined using a thermal press at 200 °C for 1 min.

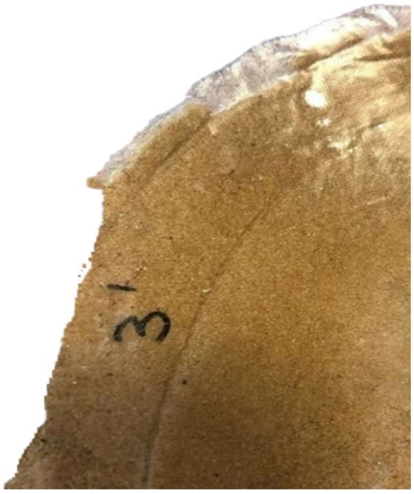


Figure 4. Compostable material produced using rice husks as a filler (formulation sample No. 8), laminated with a PLA layer.

As a result of the conducted research, a compostable composite was obtained - a mixture of heterogeneous components combined into a common structure. The resulting material belongs to the type of layered composite materials.

CONCLUSIONS

A growing problem of plastic pollution is petroleum-based polymers are characterized by a long decomposition period. The present work offers a method of using secondary raw materials of crop production as a part of compostable composite materials is proposed. As a result of research was development an advanced technology of materials` obtaining. The use of potato starch as the main component of the formulation mixture and various fillers (rice and barley husks) as reinforcing additives were considered. Two methods of gelatinization of the formulation mixture, in a water bath and in a microwave oven, as well as methods of obtaining compostable composites using an extruder, hand-rolling and press-vulcanizer were analyzed.

According to the results of tensile tests and comparative evaluation of physical and mechanical properties, the formulation with starch as a binder (17.8%), rice hulls as a reinforcing filler (34.25%) and the minimum required water content (42.96%) was accepted as optimal.

The extrusion moulding for the material under study was found to be unsatisfactory due to the low moisture resistance and low mechanical strength of the resulting material.

Thermo-press moulding is the most suitable method among those considered for producing compostable materials with the declared characteristics. The heat press for moulding blanks and laminating should provide a heating temperature of at least 200 °C for at least 5 min. The surface of the composite was laminated with a layer of polylactide to improve its thermal stability and hydrophobic properties.

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