Foods and Raw Materials, 2022, vol. 10, no. 1

E-ISSN 2310-9599
ISSN 2308-4057

Research Article
Open Access

https://doi.org/10.21603/2308-4057-2022-1-148-154
Available online at http://jfrm.ru/en

Biostability of binder-free wood and plant plastics protected with antiseptics

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Received September 22, 2021; Accepted in revised form October 12, 2021; Published online January 31, 2022

Abstract:
Introduction. Agriculture produces a lot of plant and food waste that is highly biodegradable. In order to recycle this waste and use it in the production of new materials, we need to find effective ways to increase their resistance to biodegradation. We aimed to study the biostability of binder-free wood and plant plastics, as well as to find an optimal method of their antiseptic protection.
Study objects and methods. Our objects of study were binder-free plastics based on sawdust, wheat and millet husks. To determine their biostability, we exposed them in active soil for 21 days and analyzed their physical and mechanical properties. Also, we examined the effects of several methods of antiseptic treatment on the samples' strength, water resistance, and biodegradation.
Results and discussion. All the wood- and plant-based samples showed low biostability. Exposure in active soil caused significant morphological and structural changes, as well as impaired the samples' physical and mechanical properties, especially those of the plant-based plastics. Their resistance to biodegradation was significantly determined by the type of filler or antiseptic, as well as by the method of antiseptic administration. Whether added to the press mixture or applied to the surface, the antiseptics changed the samples' physical and mechanical properties. Among the antiseptics used, copper sulfate showed the best effect when introduced directly into the sawdust press mixture. It ensured the lowest decrease in flexural strength, but increased hardness, water absorption, and swelling. The wheat- and millet-based plastics protected with copper sulfate showed an increase in strength indicators, but lower water resistance.
Conclusion. The antiseptic protection of binder-free wood and plant plastics affects a number of their physical and mechanical properties and therefore should take into account the expected conditions for their performance.

Keywords: Binder-free plant plastics, binder-free wood plastics, bioplastics, wheat husks, millet husks, biostability, biodegradation, antiseptic protection

Please cite this article in press as: Buryndin VG, Artyemov AV, Savinovskih AV, Krivonogov PS, Krivonogova AS. Biostability of binder-free wood and plant plastics protected with antiseptics. Foods and Raw Materials. 2022;10(1):148–154. https://doi.org/10.21603/2308-4057-2022-1-148-154.

INTRODUCTION

In the spotlight of current research and development are new formulations and technologies for producing plastics based on plant fibers and fillers, polymer composites combining products of traditional petrochemistry and biotechnology, biodegradable plastics, and biopolymers [1]. We can see new trends in the development of these technologies. Many of them become commercialized and acquire a wide applied significance in addition to their scientific value [2].

Recent years have witnessed a growing interest in biocomposites and bioplastics filled and reinforced with natural fibers and plant components, as well as in plant bioplastics that do not contain any products of large-scale petrochemistry [3–5]. Over the last few years, the global market of biodiversity-based plastics has had an average annual growth of 40% [3]. Largely stimulated by consumer demand, the development of bioplastics aims to improve the performance, availability, and environmental sustainability of materials and products [6]. The problem of microplastic pollution has also attracted a lot of attention recently.

The requirements for biodegradable polymers are changing: the decomposition of a polymer matrix into macro- and microscopic particles is no longer an indicator of satisfactory destruction [7]. Although actively developing abroad for several decades, bioplastic technologies are a relatively new field of research in Russia. Its President, Vladimir Putin, declared 2017 the year of ecology, which stimulated a search for low-waste and less resource-intensive production methods, as well as for recycling and waste disposal technologies to make industrial enterprises more environmentally friendly [8]. In line with this concept is the processing of wood and plant waste such as sawdust and husks of wheat, oats, buckwheat, and other crops into environmentally friendly and practical materials. Quite promising is the production of wood and plant plastics without binders [9].

Russia yearly produces significant amounts of waste suitable for recycling and processing, such as husks, oilcakes, fibers, etc. However, this waste is not widely used to produce new materials. The reasons are a lack of effective processing technologies and equipment, financial and economic aspects, and a low market interest [10].

To produce binder-free composite bioplastics based on polymers and plant fillers, wood and plant bioplastics, we need to prove their high performance properties. For example, materials with a high rate of biodegradation can be used for mulching or to make agrotechnical films, as well as disposable containers for seedlings and soil [11]. However, structural and finishing products, or reusable packaging, need to be highly resistant to various environmental factors.

For this, composites are often used whose matrix contains recycled polyethylene or polypropylene with the addition of plant components (fibers, husks, and flour). The properties of such composites are well studied [12]. For example, the resistance of wood and plant plastics is known to be determined by the biostability of the press material (its main components) and the absence of molecules that are a substrate or nutrient for soil, saprophytic micro- and macroorganisms [13, 14].

Polymer molecules can be destroyed physicochemically, through hydrolysis, under the action of acidic or alkaline media, or under the action of enzymes from fungal and bacterial cultures. Both ways of biodegradation are possible with binder-free plant and wood plastics [10]. They are mainly damaged by fungi and, to a lesser extent, by bacteria that cause rot and destroy lignin [13].

The shelf-life of household products made of binder-free wood bioplastics is estimated at 7.5 years if used at room temperature and moderate humidity [15]. Antiseptic protection is needed to maintain and improve their performance characteristics. However, materials treated with antiseptic agents change their physical, mechanical, and operational properties [16, 17]. Thus, to fully use agricultural plant waste in recycling and production of new materials, we need to find the most effective methods to increase their resistance to biodegradation.

In this regard, it seems relevant to study the biostability of binder-free wood and plant plastics based on sawdust, wheat and millet husks, as well as to find an optimal way of their antiseptic protection. Our aim was to study the biodegradation of wood (based on sawdust) and plant (based on millet or wheat husks) plastics produced without binders and treated with antiseptics.

For this, we analyzed the biostability of the samples of binder-free wood plastics and binder-free plant plastics, assessed the effect of antiseptics on their physical and mechanical characteristics, and analyzed the biostability of the samples antiseptically protected by different methods.

STUDY OBJECTS AND METHODS

Our study objects were the antiseptically treated samples of binder-free wood plastic (BF-WP) based on sawdust and binder-free plant plastic (BF-PP) based on wheat and millet husks. The samples were 2–4-mm-thick discs, 90 mm in diameter, made by pressing from raw materials containing the plant component (sawdust, wheat or millet husks). The weight of the press material was 10 g per disk. The pressing time was 10 min, pressure 124 MPa, cooling time under pressure 10 min. Some of the samples were treated with antiseptic compounds by adding them to the press material or by applying them to the finished sample after conditioning. We used a water repellent (1 g/disc), 12% $CuSO_4$ (0.6 kg/100 m^2), and a Forwood antiseptic (Raduga Coating Works, Novosibirsk) (2 g/disc). The amounts of antiseptics were based on the previous studies.

Before assessing biostability, we analyzed the physical and mechanical properties of the samples. In particular, we determined the density, flexural strength, hardness, elasticity number, compression modulus, flexural modulus, breaking stress, yield stress, water absorption, and swelling in thickness after 24 h. Then, the samples were kept in active soil for 21 days to study biostability.

The soil was prepared in accordance with State Standard 9.060-75. At the beginning of the tests, the soil extract had a pH of 7.0 and a biological activity coefficient of 0.8. The soil's microbiocenosis contained native field strains of microorganisms. After the soil exposure, the samples were analyzed for macro- and microvisual signs of biodegradation (splitting, swelling, loosening, cavities, morphological changes in the plant particles, changes in color, colonies of microorganisms, hyphae, fungal fruit bodies inside or on the surface of the sample, sliming of the surface). Then, we examined the physicomechanical parameters of those samples which were not damaged by the exposure in active soil.

RESULTS AND DISCUSSION

First, we analyzed the key physical and mechanical properties of the control samples, namely binder-free wood plastic based on sawdust (BF-WP) and binder-free plant plastics based on wheat husks (BF-PP-wheat) and millet husks (BF-PP-millet). We found that the

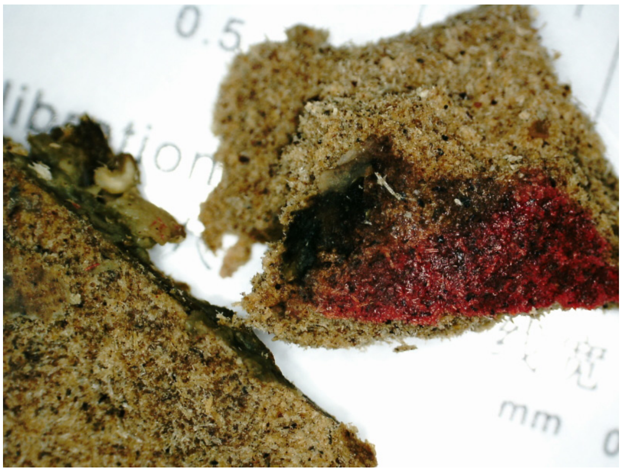

Figure 1 Pigmented colonies of microorganisms in binder-free wheat-based plant plastic after 21 days of exposure in active soil

exposure in active soil caused significant visual changes in both wood and plant samples. On average, 60% of BF-PP-millet, 58% of BF-PP-wheat, and 47% of BF-WP samples showed pronounced longitudinal and transverse splitting, edge swelling, and loosening in thickness. They also had micro- and macrocavities, especially along the edges and in the splitting areas. The defects varied from 1.5 to 5.5 mm.

All the samples featured microscopic signs of morphological changes in the plant particles: edge fibrillation, fragmentation and destruction of individual husk and sawdust particles, focal darkening, and microcavities of different size between the particles. Surface sliming and signs of mold growth were also found in all of the samples. In particular, multiple large colonies of mold fungi in different stages of maturity were present in 74% of BF-PP-millet, 85% of BF-PP-wheat, and 62% of BF-WP samples (Fig. 1).

On the whole, the visual signs of biological degradation were more pronounced in the plant-based samples. The sawdust-based samples had mainly edge and surface changes that hardly affected the middle.

The exposure in active soil had a negative effect on the physical and mechanical properties of the control samples, which were not treated with antiseptic compounds. The sawdust-based samples showed a decrease in hardness by 66%, elasticity number by 43%, compression elasticity modulus by 76%, breaking stress by 64%, and yield stress by 64% (Fig. 2).

The plant plastics based on wheat and millet husks had similar changes, namely a decrease in hardness by 62 and 70%, elasticity number by 46 and 47%, compression elasticity modulus by 73 and 80%, breaking stress by 60 and 68%, and yield stress by 60 and 68%, respectively.

The highest average of flexural strength was in the sawdust BF-WP samples (4 MPa) and the lowest was in the wheat BF-PP samples (1 MPa). Water absorption and swelling had the lowest values in the millet BF-PP samples (85%) and the highest values in the BF-WP and wheat BF-PP samples (94 and 96%, respectively).

Biostability tests showed a high biodegradability potential of all the samples. Biostability can be increased by changing the process parameters (pressing temperature, pressure, and time) [18]. However, antiseptic treatment is the main way to reduce biodegradation. An antiseptic component can be either added to the raw mixture or applied to the finished product. Thus, antiseptic treatment is a prerequisite for using binder-free wood and plant plastics in highly bioactive conditions, i.e., in an aggressive microbial destructive environment.

At the next stage, we treated the experimental plastics with antiseptics by adding them to the press material or applying to the surface to protect the

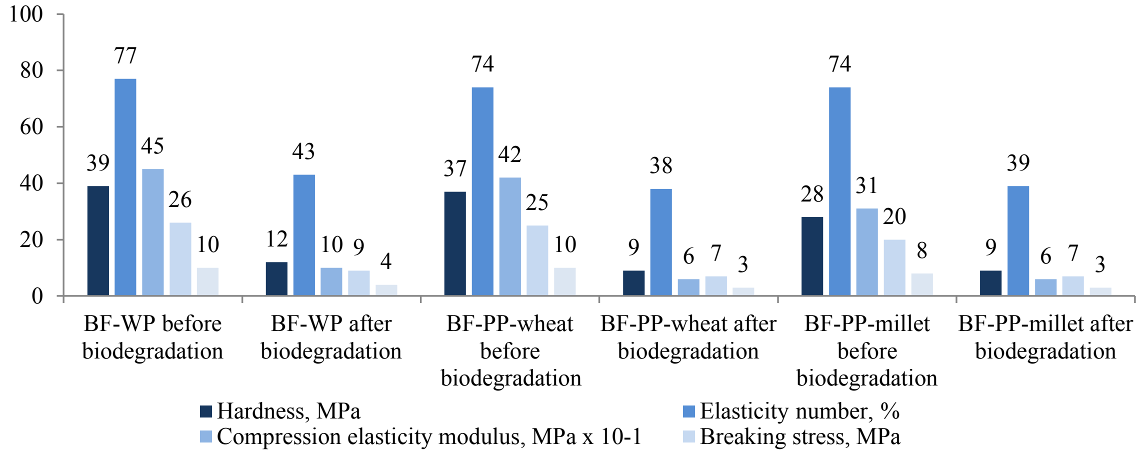

Figure 2 Changes in physical and mechanical properties of binder-free wood and plant plastics BF-WP and before and after biodegradation in active soil

Table 1 Physical and mechanical properties of binder-free wood plastics protected with antiseptic coating (biostability tests)

Physical and mechanical properties	Control			Antiseptic coating								
				Water repellent			Copper sulfate			Forwood antiseptic		
	Week											
	1	2	3	1	2	3	1	2	3	1	2	3
Flexural strength, MPa	3.4	2.6	2.0	3.5	2.8	2.6	3.2	2.8	2.7	3.2	2.3	2.2
Hardness, MPa	8.6	8.6	8.6	17.2	9.1	9.0	9.0	8.8	8.9	9.2	9.0	8.9
Elasticity number, %	40	39	39	65	38	37	41	41	41	51	42	37
Compression elasticity modulus, MPa	62	58	58	156	64	63	63	61	61	64	62	62
Breaking stress, MPa	6.4	6.4	6.4	12.3	6.8	6.7	6.7	6.6	6.7	6.8	6.7	6.7
Yield stress, MPa	2.6	2.6	2.6	4.9	2.7	2.7	2.7	2.6	2.7	2.7	2.7	2.7
Water absorption in 24 h, %	82	82	95	49	55	56	67	72	71	54	76	84
Swelling in thickness in 24 h, %	6.1	6.8	8.4	3.7	6.5	7.5	5.5	7.0	7.4	5.6	7.6	8.1

material from biodegradation, improve its biostability, and reduce its biodegradation potential. The samples' physical and mechanical properties were analyzed before and after biostability tests. We found that these properties were affected by the type and method of antiseptic administration.

The BF-WP samples had the worst indicators when a water repellent was introduced directly into the press material. In particular, there was an average decrease in flexural strength by 49%, hardness by 14%, water absorption by 30% (after 24 h), and swelling in thickness by 1.5% (after 24 h).

This might be explained by the disturbed formation of supramolecular bonds between the mixture particles during pressing. Lignin was present in the liquid phase of the mixture and the water repellent distributed it on the surface of the particles, providing them with hydrophobic properties. Thus, this modification became a structural and mechanical factor that interfered into the formation of bonds between the particles. However, when applied to the surface of the BF-WP samples, the water repellent improved their physical and mechanical properties by an average of 1–10%.

The best indicators were found in those BF-WP samples which were protected with copper sulfate introduced into the press material. They had the highest values of hardness, compression elasticity modulus, and breaking stress compared to all the other experimental (protected) and control (unprotected) samples.

A similar picture was observed in the binder-free plant plastics. Introduced into the press mixture, the water repellant caused a sharp deterioration in flexural strength and water absorption (by 10 and 11%, respectively). When applied to the surface, it improved these properties by 10 and 14%, respectively. Copper sulfate that was introduced directly into the press mixture increased the strength indicators (flexural strength by 14%, hardness by 49%), but reduced water resistance (water absorption rose by 23% and swelling by 28%). These effects must be taken into account when formulating binder-free, antiseptically protected plastics based on wood and plant materials.

The experimental BF-WP and BF-PP samples were exposed in active soil for 21 days and then tested for biostability. Our analysis of the physical and mechanical properties of the control (unprotected) and experimental BF-WP samples showed a significant decrease in strength indicators. The greatest decrease in flexural strength (by 39%) was found in the controls. This indicator fell by 29% in the samples treated with

Table 2 Physical and mechanical properties of binder-free wood plastics protected with an antiseptic introduced into the press mixture (biostability tests)

Physical and mechanical properties	Control			Antiseptic introduced into the press mixture					
				Water repellent			Copper sulfate		
	Week								
	1	2	3	1	2	3	1	2	3
Flexural strength, MPa	3.4	2.6	2.0	1.1	1.1	0.8	4.7	3.9	3.7
Hardness, MPa	8.6	8.6	8.6	8.9	8.4	8.4	16.2	14.1	10.3
Elasticity number, %	40	39	39	50	45	41	48	34	33
Compression elasticity modulus, MPa	62	58	58	61	57	57	146	121	76
Breaking stress, MPa	6.4	6.4	6.4	6.6	6.3	6.3	11.5	10.1	7.6
Yield stress, MPa	2.6	2.6	2.6	2.7	2.5	2.5	4.6	4.1	3.1
Water absorption in 24 h, %	82	82	95	110	115	115	44	47	51
Swelling in thickness in 24 h, %	6.1	6.8	8.4	7.5	7.7	9.8	4.2	4.5	4.5

Table 3 Physical and mechanical properties of binder-free plant plastics protected with antiseptic coating (biostability tests)

Physical and mechanical properties	Control			Antiseptic coating								
				Water repellent			Copper sulfate			Commercial antiseptic		
	Week											
	1	2	3	1	2	3	1	2	3	1	2	3
Flexural strength, MPa	2.0	1.8	1.3	2.5	1.7	1.1	2.1	2.0	1.6	1.6	1.2	1.0
Hardness, MPa	8.7	8.7	8.3	9.1	8.7	8.7	8.8	8.6	8.5	8.9	8.7	8.4
Elasticity number, %	36	36	41	33	38	39	39	40	40	36	38	40
Compression elasticity modulus, MPa	60	59	56	64	60	59	60	59	58	62	60	57
Breaking stress, MPa	6.5	6.4	6.2	6.8	6.5	6.5	6.6	6.4	6.4	6.7	6.5	6.3
Yield stress, MPa	2.6	2.6	2.5	2.7	2.6	2.6	2.6	2.6	2.6	2.7	2.6	2.5
Water absorption in 24 h, %	89	113	158	90	121	151	103	119	151	88	101	122
Swelling in thickness in 24 h, %	7.5	7.9	9.6	5.2	5.7	7.8	5.2	6.0	7.5	7.4	7.5	7.9

Table 4 Physical and mechanical properties of binder-free plant plastics protected with an antiseptic introduced into the press mixture (biostability tests)

Physical and mechanical properties	Control			Antiseptic introduced into the press mixture					
				Water repellent			Copper sulfate		
	Week								
	1	2	3	1	2	3	1	2	3
Flexural strength, MPa	2.0	1.8	1.3	0.4	0.3	0.3	2.0	0.6	0.3
Hardness, MPa	8.7	8.7	8.3	8.9	8.9	8.9	8.9	8.8	8.7
Elasticity number, %	36	36	41	37	38	38	34	38	38
Compression elasticity modulus, MPa	60	59	56	62	61	61	62	60	60
Breaking stress, MPa	6.5	6.4	6.2	6.7	6.6	6.6	6.7	6.6	6.5
Yield stress, MPa	2.6	2.6	2.5	2.7	2.7	2.7	2.7	2.6	2.6
Water absorption in 24 h, %	89	113	158	161	203	228	135	135	171
Swelling in thickness in 24 h, %	7.5	7.9	9.6	5.1	5.2	5.3	5.3	5.4	5.6

the Forwood antiseptic and by 26% in the samples with a water repellent introduced into the press mixture (Tables 1 and 2).

Flexural strength had the smallest losses in the samples protected with copper sulfate, namely 15% for the coated sample and 21% for the sample with a modified press mixture.

Hardness was the highest in the wood plastics treated with the water repellent and those with the copper sulfate-modified press mixture, namely 17.2 and 16.2 MPa, respectively, on the eighth day of exposure in active soil. However, it was these samples that had the greatest loss of hardness by the end of the biostability test, by 48 and 36%, respectively. Yet, this indicator remained the highest in the samples with added copper sulfate (10.3 MPa).

Water absorption had the highest values in the samples with an added water repellant, averaging 115% after three weeks of exposure. The lowest values were in the samples with added copper sulfate, namely 51% by the end of the tests (a loss of 16%).

The plant plastics also showed changes in their physical and mechanical parameters. The smallest loss (24%) of flexural strength over three weeks of exposure in active soil was found in the samples with an added water repellent, although they had one of the lowest values (0.4 MPa) in the first week. On the eighth day, this indicator was the highest in the samples coated with a water repellent (2.5 MPa), decreasing by 53% to 1.1 MPa by the end of the test (Table 3). The hardness indicator decreased in all the plant samples within three weeks of exposure in the range of 1–6%.

Daily water absorption was the highest in the samples with an added water repellent, amounting to 228% after three weeks of exposure in active soil, with a 42% decrease of water absorption. By the end of the biostability tests, the lowest water absorption was in the samples coated with the Forwood antiseptic (122%).

CONCLUSION

Our study showed high biodegradation and low biostability of the binder-free wood and plant plastics based on sawdust, and wheat and millet husks. Therefore, antiseptic protection is required to improve their performance. Their exposure in a bioactive environment caused some morphological and structural changes, as well as affected their physical and mechanical properties.

We found that the plant-based plastics underwent a more pronounced degradation in active soil than the sawdust-based plastics. According to our results, the samples' resistance to biodegradation was determined by such process parameters as the type of filler and antiseptic, as well as the method of antiseptic administration.

We treated the plastics with three types of antiseptic (water repellent, copper sulfate, and Forwood) by adding them to the press mixture or applying them to the surface. Both methods changed the initial properties of the samples. When used as a coating, the water repellent improved the samples' physical and mechanical properties. When added to the press mixture, however, it significantly impaired their strength and water resistance.

Copper sulfate showed the best effect among those antiseptics introduced into the press mixture. It decreased the flexural strength of the sawdust-based samples by 5% and increased their hardness, water absorption, and swelling. The plant-based samples with added copper sulfate showed better strength indicators, but lower water resistance. Thus, the antiseptic treatment of binder-free plastics based on wood or plants affects a number of their key physical and mechanical properties and should be administered with regard to expected performance conditions.

CONTRIBUTION

All the authors were equally involved in developing the research concept, obtaining and analyzing the data, and writing the manuscript.

CONFLICT OF INTEREST

The authors declare that there is no conflict of interest.

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