

## Article

# The Possibilities of Using Poly(3-hydroxybutyrate-co-3-hydroxyvalerate) PHBV in the Production of Wood–Polymer Composites

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**Abstract:** In this study, polymer biocomposites based on poly(3-hydroxybutyrate-co-3-hydroxyvalerate) PHBV biopolymers with Arbocel C350 SR wood fiber filler with mass contents of 15%, 30%, and 45% were described. Samples for testing were produced using the injection molding process. The shrinkage of the produced composites was determined, as well as the basic mechanical properties on the basis of the uniaxial static tensile test, hardness, and impact tensile test. The dimensional stability of samples was subject to temperature and humidity in the water absorption test. This research was carried out in terms of the problems with composite processing and use of products. This paper contains many remarks and conclusions regarding the processing and exploitation of the tested products, which can be extended to a larger range of cellulose fillers. It was found that it was possible to produce the tested type of composites with a content of up to 45 wt. of filler. However, the mechanical properties of the tested composites made it possible to use them for the production of selected products. These conclusions allow for conducting future research toward the effective use of WPC composites with a PHBV matrix and fibrous fillers of natural origin.

**Keywords:** PHBV; WPC; PHBV–wood filler biocomposites; polymer composites; injection molding process



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## 1. Introduction

Polyhydroxyalkanoate (PHA) plastics have fascinated scientists and engineers due to their unique properties and industrial potential. Primarily known as biodegradable, biocompatible, and thermoplastic polyesters, PHAs present a wide spectrum of applications, from packaging, through medicine, to agriculture [1]. PHA is a family of polyesters produced naturally by bacteria as a backup material. These plastics are characterized by high biodegradability, which means that, under the influence of microorganisms, they are able to completely decompose into simple chemical compounds, such as water and carbon dioxide. In addition, PHAs are biocompatible, which makes them excellent candidates for medical applications, such as surgical sutures or drug-release systems [2–4]. PHA processing can be performed by various technologies that are similar to those used for conventional thermoplastics. The most popular methods include injection, foil stamping, extrusion, blow molding, and thermoforming. In this way, PHAs can be easily introduced into existing production chains [5,6]. The production of PHA on an industrial scale often involves a biotechnological process in which microorganisms, such as bacteria, ferment various substrates (e.g., starch or glucose) and accumulate PHA in their cells. Then, these polymers are extracted and processed into various products [7,8].

Poly(3-hydroxybutyric acid) is a polymer belonging to the group of polyhydroxyalkanoates and, in some aspects, is similar to polypropylene in terms of physical properties [9]. Nevertheless, its tendency to form large crystalline domains in the form of spherulites

makes it brittle, which may be due to the exceptional purity of the biopolymer. This specificity of PHB is interesting for microbiological studies of spherulites, but at the same time, it is a significant limitation in the context of the commercial use of this homopolymer [10,11]. One way to improve the mechanical properties of PHB is to add poly(3-hydroxyvaleric acid) (HV or PHV) to the PHB chain, which allows for obtaining PHBV. It has been noted that the PHB and HV units in the PHBV copolymers have a statistically random distribution. Thus, such copolymerization reduces the crystallinity [12–14]. Compared with PHB, PHBV exhibits lower stiffness and brittleness, as well as higher tensile strength and higher elongation at break. Additionally, by varying the amount of HV in the polymer chain, the melting point can be significantly lowered while also increasing the difference between the melting point and the onset of thermal decomposition. This, in turn, allows the processing window to be improved [15].

Owing to its biodegradability and easy processability, PHBV is still being modified and commercialized as the main substitute for non-biodegradable polymeric materials. The similarity of some of its mechanical properties to those of polyolefins indicates that it can be a substitute for polymers from this group [16]. HV can be produced by adding propionic acid as a nutrient feed to the bacteria. The molar fraction of HV in the PHBV polymer is limited due to the toxicity of propionic acid to the *Ralstonia eutropha* bacterium. PHBV has also been produced biologically using microorganisms, such as *Rhodococcus* [17] and *Chromobacterium violaceum* [18], as well as using the recombinant *Ralstonia eutropha* bacterium [19]. PHBV is the first PHA biopolymer with properties that can be changed by controlling the content of the second monomer [20,21]. The share of HV in PHBV significantly affects the properties of the polymer—the melting point decreases from the initial temperature of 180 °C to only 75 °C (at approx. 40% molar share of HV). Then, as the HV content continues to increase, the melting point also increases, and at 95% molar share of HV, the melting point is 108 °C. It has also been noticed that the change in the content of HV in the polymer regulates the rate of crystallization and impacts strength, Young's modulus, and tensile strength [22–24].

Wood fillers are an increasingly popular solution in the plastics industry. The use of these fillers in the polymer matrix is intended to improve some mechanical and thermal properties and to produce ecological composite materials. Adding wood fillers to the polymer matrix can improve the strength, stiffness, and hardness of the composite material. Wood fibers in particular are known for their excellent mechanical properties and can significantly strengthen the polymer structure [25]. In addition, this type of filler can improve the thermal stability of composite materials. This acts as a thermal barrier that protects the polymer against high temperatures and reduces its tendency for deformation and degradation [26,27]. The use of wood fillers can contribute to increasing the biodegradability of composite materials. Polymers combined with them could be more environmentally friendly, as wood is a renewable resource and decomposes more easily than traditional polymers. They are also attractively priced compared with other fillers of natural origin, e.g., flax, hemp, etc. [28,29].

Wood–polymer composites (WPC) can be processed using the traditional injection molding process [30,31]. These composites can also be extruded [32,33]. This processing technique is widely used in the packaging or construction industry, as well as other sectors. The use of wood fillers in the polymer matrix is a promising direction for the development of composites and will contribute to a greener and more sustainable future.

Nowadays, new materials are being sought. These are so-called double-green polymers to replace currently commercially used petrochemical-derived polymers. Unfortunately, as in the case of PHBV, the major barrier here is the production cost of these types of bioplastics, which are not attractively priced for companies specializing in plastics processing. The production of composites consisting of a wood filler and PHBV can be an alternative. The issues of the production, processing, and evaluation of some of the mechanical processing and functional properties of such composites are included in this work.

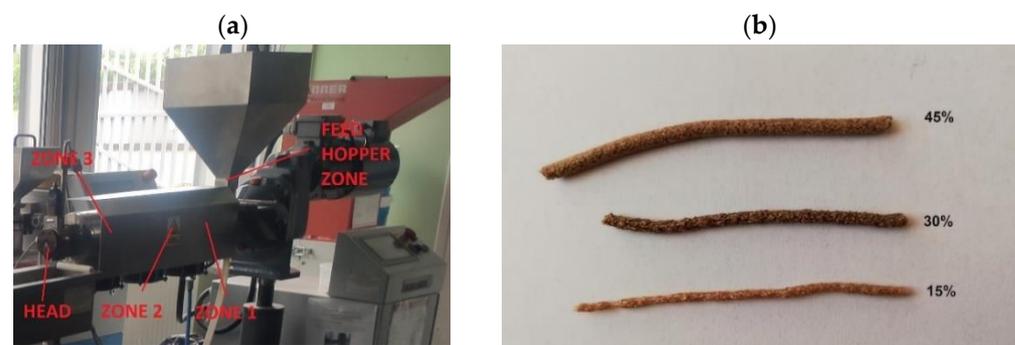
## 2. Materials and Methods

### 2.1. Research Materials

WPC composites with different contents of wood fibers were produced and examined in this work. Wood fibers under the trade name Arbocel C350 SR produced by J. Rettenmaier & Söhne GmbH Corporation (Rosenberg, Germany) were used as the filler. The matrix of the biocomposite was the PHBV ENMAT Y1000 biopolymer from NaturePlast (Mondeville, France).

### 2.2. Preparation of Biocomposites and Specimens

The mixture of PHBV biopolymer and wood fibers was plasticized in an extruder. In the extrusion process, the ZAMAK EHP-25 (produced by ZAMAK Mercator company, Skawina, Poland) single-screw extruder was used. The mass shares of the wood filler were 15%, 30%, and 45%, respectively. The extrusion process was carried out at a screw speed of 80 rpm. Due to processing difficulties, different temperature profiles of the plasticization unit of the extruder were used for each percentage share (Tables 1 and 2). In order to prepare pellets from biocomposites, a profile with a circular cross-section was extruded (Figure 1). The densities of the WPC composites in the solid state were as follows: 1250 kg/m<sup>3</sup> (for 5% wt. of the filler), 1251 kg/m<sup>3</sup> (for 30% wt. of the filler), and 1252 kg/m<sup>3</sup> (for 45% wt. of the filler).



**Figure 1.** (a) Extruder with marked heating zones, (b) extrudates produced for different contents of wood fibers.

**Table 1.** WPC (PHBV + Arbocel C350 SR) composites extrusion pressures.

Mass content of wood fibers (%)	15	30	45
Extrusion pressure (MPa)	0.3	0.49	0.54

**Table 2.** Processing parameters of plasticization unit (see Figure 1a).

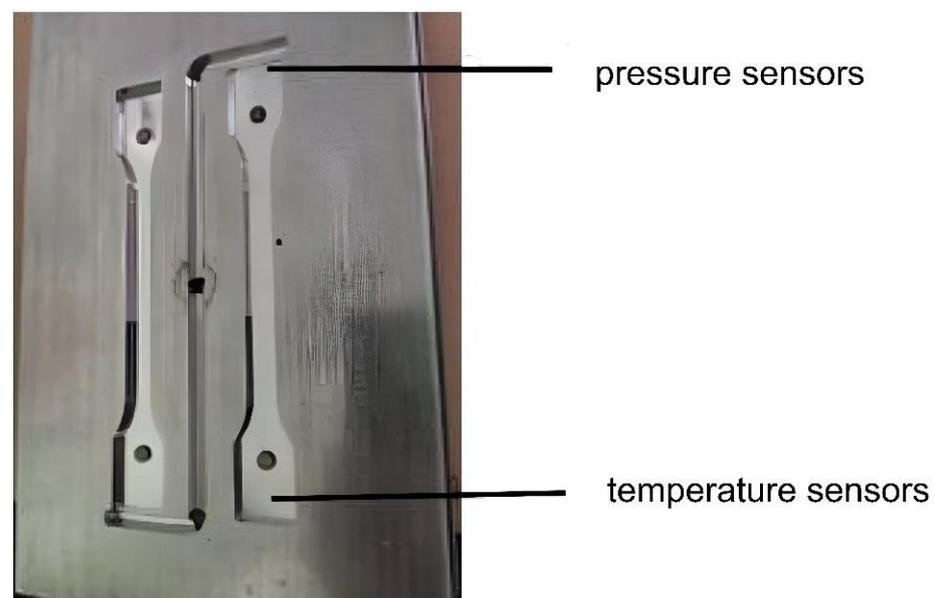
Mass Content of Wood Fibers (%)	Temperature (°C)			
	Head	Zone 3	Zone 2	Zone 1
15	164	162	154	145
30	175	170	160	150
45	180	175	165	155

A biocomposite was produced for each amount of filler. It was found that, with increasing fiber content, for the same processing parameters, the cross-sectional diameter of the extrudate increased (Figure 1b).

In the most commonly used granulation method, the hot material, during the cooling stage, is cooled down in a cooling bath. During this process, the extrudate intensively absorbs water. No such bath was used in the present study. The extrudate was dried in the

open air and the granulate was obtained in the process of grinding it using a low-speed mill, type C17, manufactured by WANNER (Wertheim, Germany).

The obtained granulate was dried in a DZ-2BC vacuum dryer (produced by Chemland company, Szczecin Stargard, Poland) before the injection process. Drying was performed for 3 h at 80 °C. The dried granulate was used to inject the samples. A Dr BOY injection molding machine type 55 E (produced by BOY Maschines Inc., Exton, PA, USA) was used for the injection process. A Priamus system with two pressure sensors (marked P1 and P2) and temperature sensors (marked T1 and T2) mounted at the beginning and at the end of the injection mold cavity was used to record and analyze the pressure and temperature in the cavities of the injection mold in order to control the processing parameters. (Figure 2). During the production of samples from each type of composite, 30 production cycles were recorded. In the injection molding process, 3 sets of samples containing 15%, 30%, and 45% wood filler were obtained.



**Figure 2.** The cavity insert with mounted temperature and pressure sensors.

The processing parameters of the injection molding process are listed in Tables 3 and 4.

**Table 3.** Selected processing parameters of the injection molding of the PHBV–wood fiber composite for the following phases: injection, packing, and plasticization.

Filling Phase			
Max. pressure [MPa]	100	Velocity [mm/s]	400
Packing phase			
Pressure [MPa]	33	Packing time [s]	30
Plastification phase			
Pressure [MPa]	3.5	Velocity [mm/s]	130

**Table 4.** Mold temperature depending on the mass content of wood fibers in the composite.

Mass Content of Wood Fibers [%]	Mold Temperature [°C]
15	65
30	85
45	90

Different injection mold temperatures were used, depending on the filler content of the composite. The mold temperature is listed in Table 4.

### 2.3. Testing Methods

Many tests were performed as a part of the assessment of the mechanical, processing, and utility properties of the tested WPC composite.

Shrinkage tests were performed in accordance with the following standard: ISO 294-4 [34]. These tests were performed after 3 h. The dimensions of seven samples were measured for each mass content of the used filler.

The evaluation of basic mechanical properties for the produced composites was performed using a ZWICK/ROELL Z030 testing machine (produced by Zwick Roell, Ulm, Germany). The test was carried out in accordance with the standard ISO 527-1 [35]. Each series of samples consisted of 7 pieces for subsequent statistical analysis.

The hardness of the samples was measured using a ZWICK 3106 hardness tester (produced by Zwick Roell, Ulm, Germany). The test was performed in accordance with the standard ISO 2039-1 [36]. From the WPC composite molded pieces with different contents of wood fibers, seven samples were selected to test each filler content. The measurement was carried out for the samples with dog-bone geometry. Six measurements were performed on each sample, three for two areas: area I—near the gate—the area that includes points 1, 2, and 3; and area II—at the end of the plastic flow path in the mold cavity—the area that includes points 4, 5, and 6 (Figure 3).

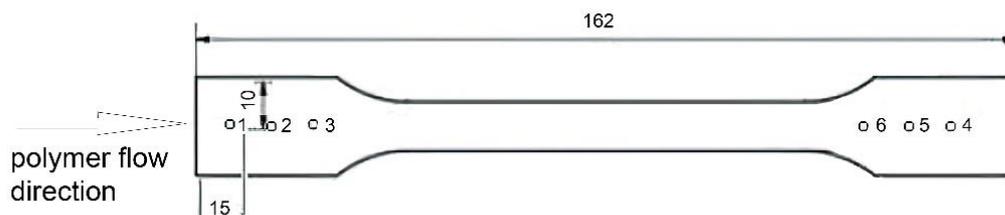


Figure 3. Hardness measurement areas.

Biocomposite samples were also tested in the impact tensile test. The impact tensile strength was determined in accordance with the PN-EN ISO 8256 standard [37]. For this purpose, the CAEST 9050 pendulum hammer (produced by Instron Inc. Europe, Buckinghamshire, UK) was used. Their geometry was modified in accordance with the standard. The notch was milled for all sample groups. Each series of samples consisted of 7 pieces for subsequent statistical analysis.

Two tests were performed to assess the utility properties of the obtained composite: dimensional stability and water absorption tests.

In the temperature dimensional stability test, the samples as presented in Figure 3 were initially placed in a freezer at  $-6\text{ }^{\circ}\text{C}$  and stored for 1.5 h until the temperature stabilized. After removal, they were immediately measured. Subsequent measurements were performed after the thermal stabilization of the samples to the desired temperature for 1 h in a Chemland vacuum dryer, type DZ-2BC (produced by Chemland company, Szczecin Stargard, Poland).

The water absorption test was carried out in accordance with the standard ISO 62 [38]. The samples were placed in a container with water and the color changes were observed. These changes were evaluated organoleptically. Before the measurements, the samples were dried after taking them out of the water and left for 5 min on a dry surface in order to dry. The samples were measured and weighted.

## 3. Results

During the production of the tested WPC biocomposites, an increase in the extrudate diameter was found with the amount of wood fiber (Figure 1b), compared with the nominal diameter of the extrudate of 2 mm. The average values of extrudate diameter are presented

in Table 5. During the extrusion process, the diameter of the extrudate for the individual filler contents was not characterized by a large dispersion (8% for the filler content of 15% by weight, 6% for the filler content of 30% by weight, and 5% for the filler content of 45% by weight).

**Table 5.** Changes in extrudate diameter for different mass contents of filler.

Mass content of wood fibers	15%	30%	45%
Diameter (mm)	2.06	2.69	3.34

The effect of increasing the cross-section of the extrudate with the share of the filler was determined using dependence 1 (Table 6). The plastics leaving the extruder head swelled. The extrudate diameter increase factor  $\beta$  was calculated from relationship (1):

$$\beta = \frac{D_w}{d_k} \tag{1}$$

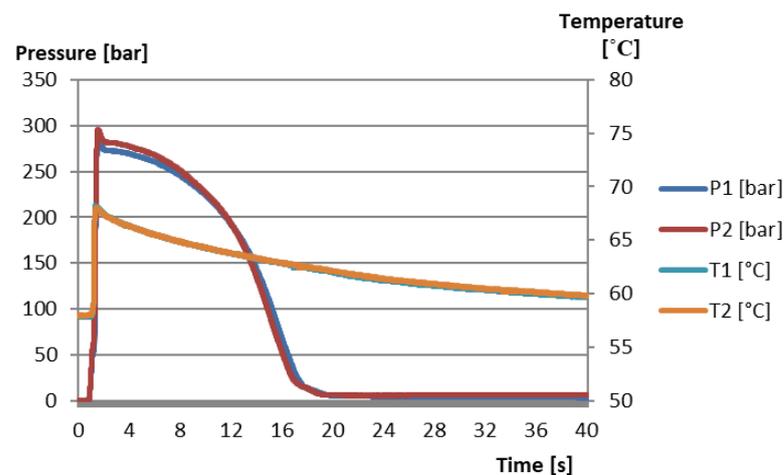
where:

$D_w$ —extrudate diameter;  
 $d_k$ —head diameter.

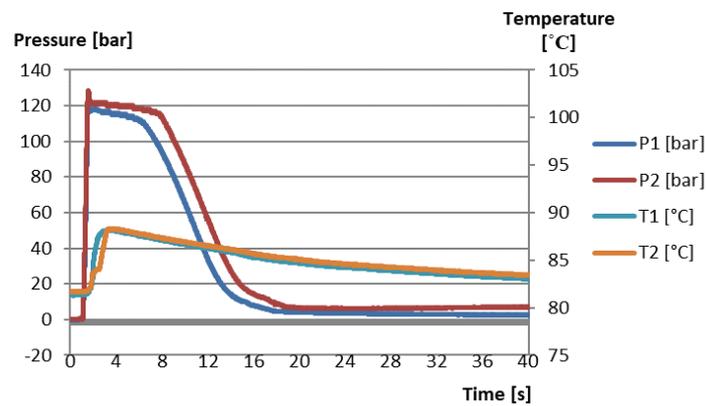
**Table 6.** Changes in the diameter of the extrudate for composites PHBV–wood fiber with different contents of wood fibers.

Mass Content of Wood Fibers	$\beta$
15%	1.03
30%	1.34
45%	1.67

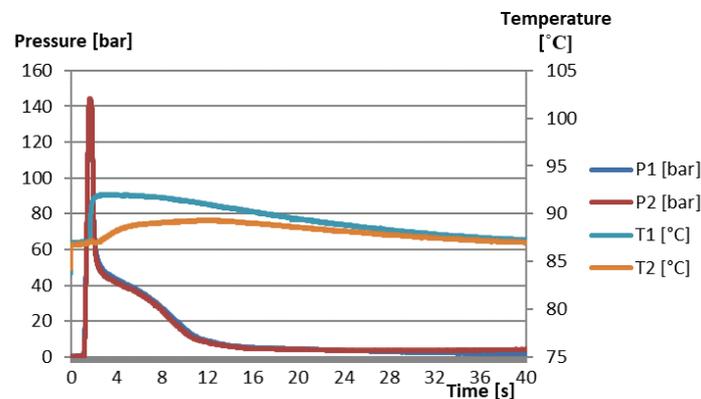
Representative characteristics of the injection molding process, i.e., pressure and temperature graphs over time, recorded in mold cavities using the Priamus system, are shown in Figures 4–6.



**Figure 4.** Exemplary courses of pressure and temperature changes of the WPC composite with 15% wood filler content in the injection mold cavities (cycle 10/30).



**Figure 5.** Exemplary courses of pressure and temperature changes of the WPC composite with 15% wood filler content in the injection mold cavities (cycle 25/30).



**Figure 6.** Exemplary courses of pressure and temperature changes of the WPC composite with 15% wood filler content in the injection mold cavities (cycle 30/30).

Most of the specimens were shaped correctly, but defects appeared in the injection molding process of the composite with a mass content of 45% wood fibers. The problem was the incomplete filling of the mold cavity. The packing time was then modified, which improved the quality of the manufactured molded pieces. The defects that were observed in the form of incomplete filling of the cavities and the defects in the shapes of the samples are shown in Figure 7.



**Figure 7.** Examples of molded pieces' defects for 45% by weight of wood fibers (unfilled areas are marked).

**Primary shrinkage assessment**

The measurement results of the primary shrinkage assessment are presented in Table 7.

**Table 7.** Primary shrinkage values in the following directions: longitudinal  $S_{II}$ , transverse  $S_{\perp}$ , thickness  $S_g$ , volumetric  $S_v$ , SD—standard deviation.

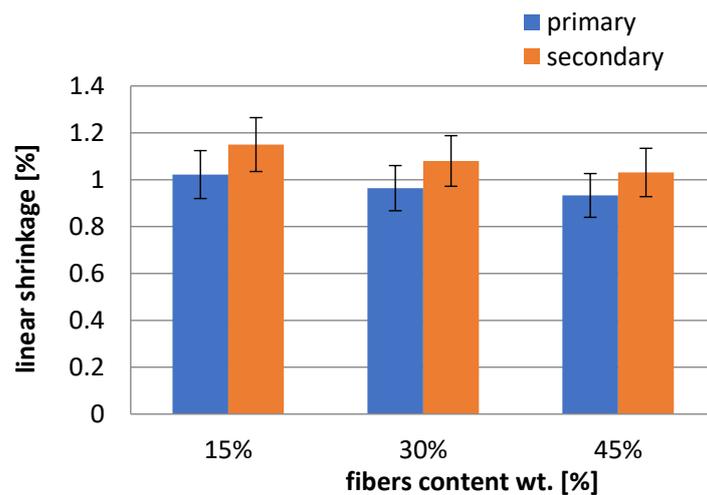
Filler Content	$S_{II}$ (%)	SD (%)	$S_{\perp}$ (%)	SD (%)	$S_g$ (%)	SD (%)	$S_v$ (%)
15%	1.022	0.091	2.127	0.17	2.857	0.371	1.046
30%	0.964	0.077	2.54	0.254	3.312	0.331	0.872
45%	0.933	0.083	2.647	0.397	3.167	0.253	0.76

**Secondary shrinkage assessment**

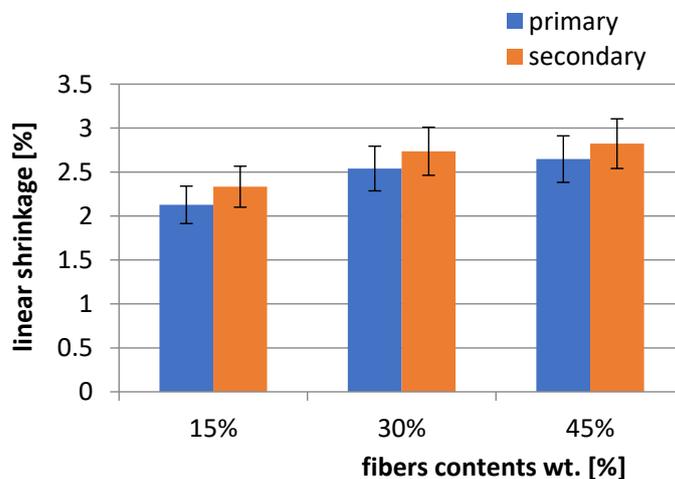
After 24 h, the samples were measured again. Based on the measurements, the values of linear and volumetric shrinkage were calculated. The shrinkage values, both primary and secondary, depending on the fiber content are presented in Table 8 and Figures 8–11.

**Table 8.** Secondary shrinkage values in the following directions: longitudinal  $S_{II}$ , transverse  $S_{\perp}$  and in thickness  $S_g$ , volumetric  $S_v$ , SD—standard deviation.

Filler Content	$S_{II}$ (%)	SD (%)	$S_{\perp}$ (%)	SD (%)	$S_g$ (%)	SD (%)	$S_v$ (%)
15%	1.150	0.112	2.333	0.186	3.238	0.356	1.055
30%	1.080	0.086	2.736	0.328	3.619	0.370	0.854
45%	1.031	0.092	2.823	0.282	3.500	0.035	0.724



**Figure 8.** Longitudinal shrinkage values for different filler contents.



**Figure 9.** Transverse shrinkage values for different filler contents.

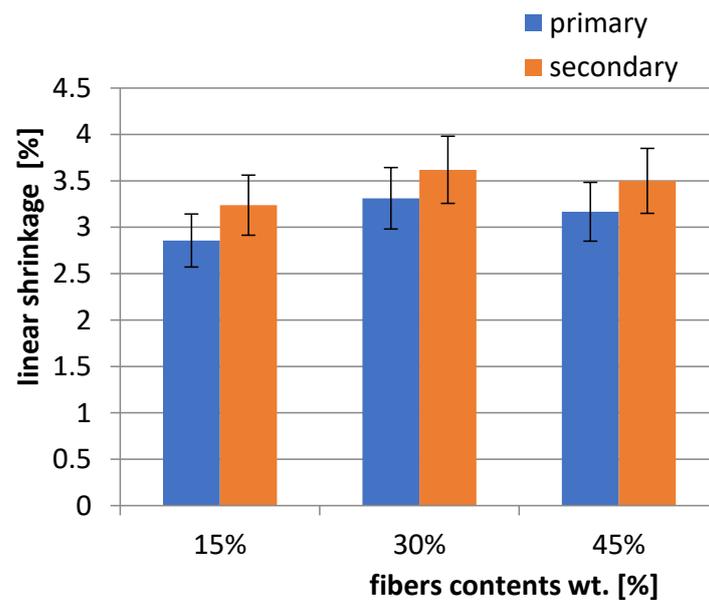


Figure 10. Thickness shrinkage values for different filler contents.

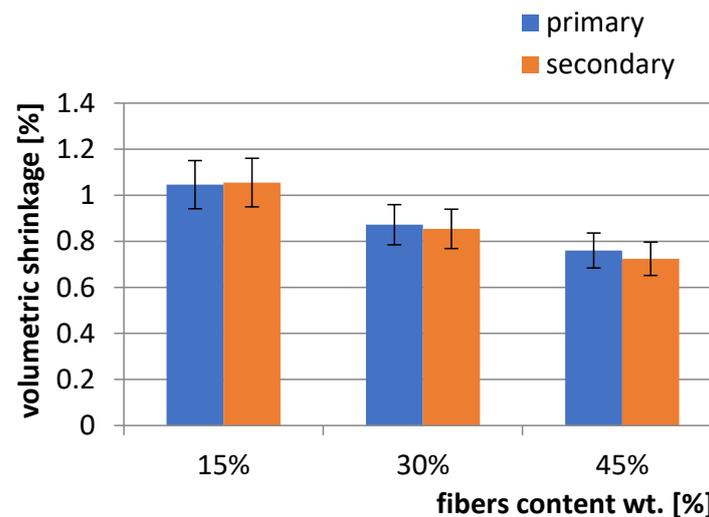


Figure 11. Volumetric shrinkage values for different filler contents.

### Hardness test

The measurements of hardness in sample areas 1, 2, and 3 (Figure 3) did not allow the determination of the trend of changes in the hardness values for all used contents of filler. In these areas, it may be due to the longest flow of polymer material during filling and disturbed arrangement of wood fibers in the area close to the gate. Samples containing 30% (by weight) of wood fibers showed the highest hardness. WPC composites with filler contents of 15% and 45% by weight had similar hardnesses. Measurements were also performed for points 4, 5, and 6. For these places, a tendency to increase the hardness was found with the distance from the edge of the sample. The results of hardness measurements are presented in the graphs (Figure 12).

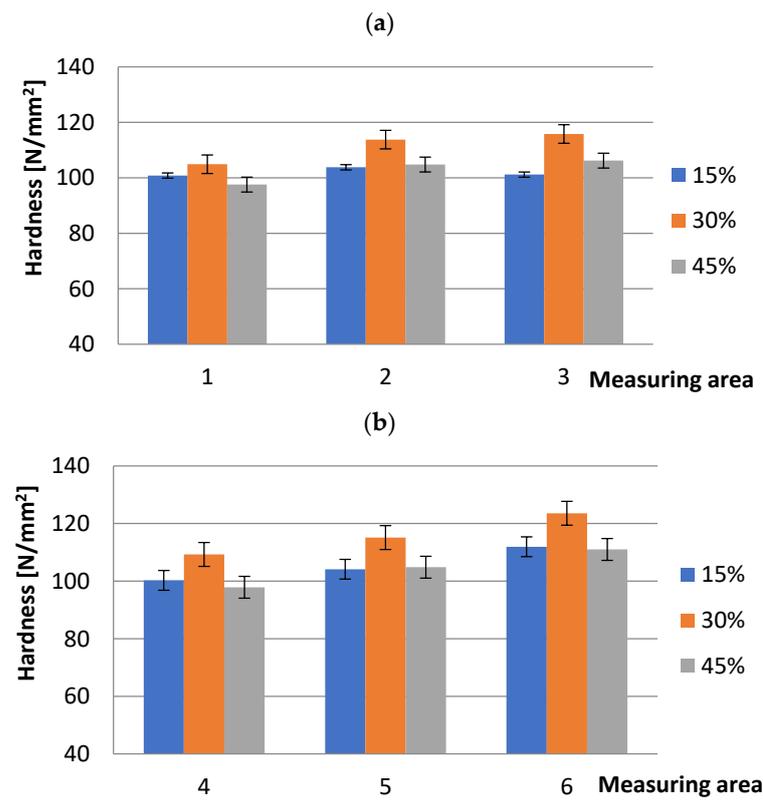


Figure 12. Hardness of samples with different contents of wood fiber (a) for area I (points 1, 2, and 3) and (b) for area II (points 4, 5, and 6).

**Mechanical properties—static tensile test**

In this test, the basic mechanical properties of the obtained composites were determined. The relationship between stress and strain for the tested composites is shown in Figure 13. The specific mechanical properties of PHBV–wood fiber composites are presented in Table 9.

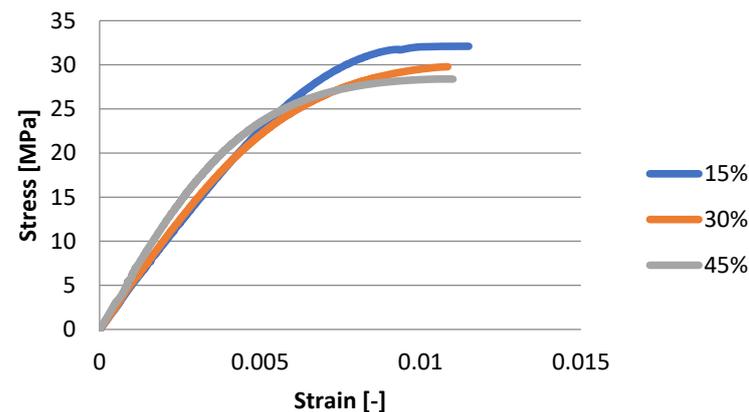
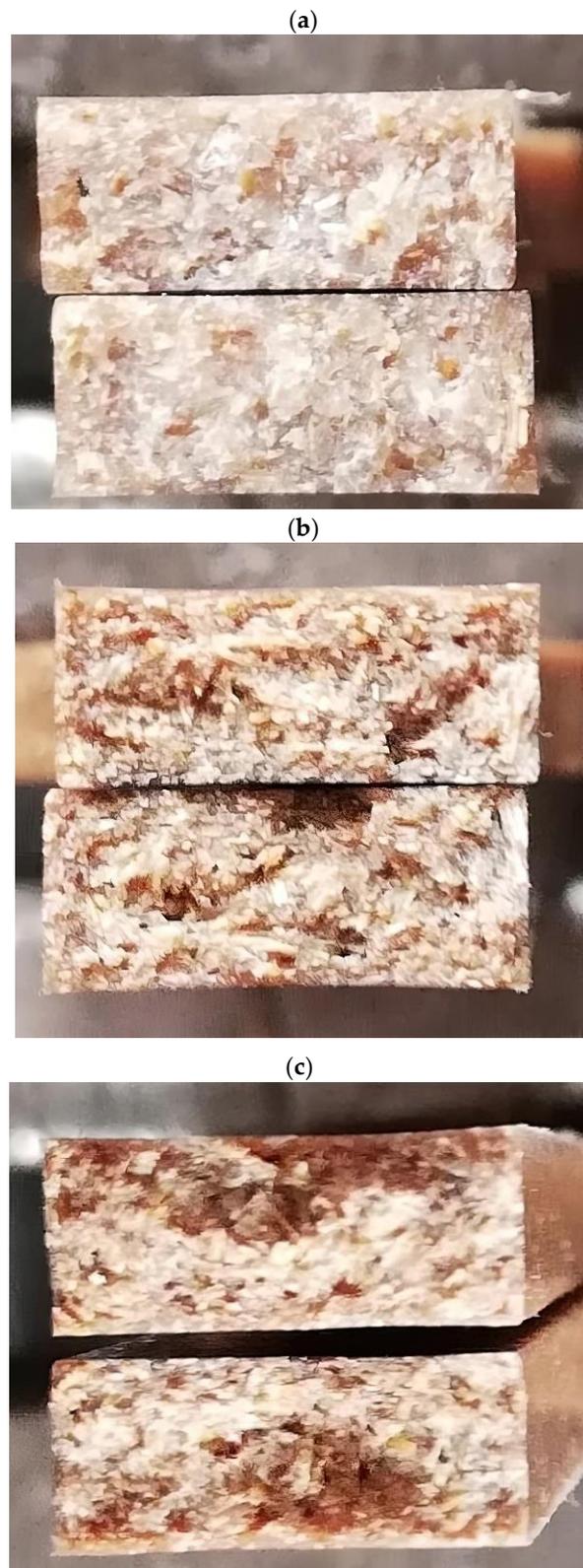


Figure 13. Stress–strain characteristics for the tested wood fiber contents in PHBV–wood fiber composites.

Table 9. Chosen mechanical properties of PHBV–wood fiber composites.

Fiber Amount by Weight (%)	$E_t$ (MPa)	$\sigma_M$ (MPa)	$\epsilon_B$ (%)
15	5494	31.9	1.199
30	6044	29.9	1.072
45	6361	27.9	1.008

The fractures obtained after the destruction of the samples with different contents of wood filler in the uniaxial tensile test were definitely different. They are presented in Figure 14a–c).



**Figure 14.** View of the surface after destruction of PHBV–wood fiber composite samples for different filler contents by weight: (a) 15%, (b) 30%, and (c) 45%.

The fracture of the sample containing 15% wood fibers by weight was the lightest in terms of color. Individual fibers were hardly visible because the sample contained a lot of the PHBV matrix. This destruction surface was also characterized by the greatest homogeneity of the structure. On the other hand, the destruction surface of the sample containing 45% wood filler by weight was distinguished by the darkest color. Wood fibers were more visible, and they had the least homogeneous structure. Densities of wood fibers formed in certain areas, which resulted in the deterioration of some properties. The destruction surface of the WPC composite with 30% wood fibers by weight had the most evenly distributed fibers.

**Impact tensile test**

The purpose of the test was to determine the changes in impact tensile strength depending on the wood fiber content. The dependence of the impact tensile strength on the wood filler content is shown in Figure 15.

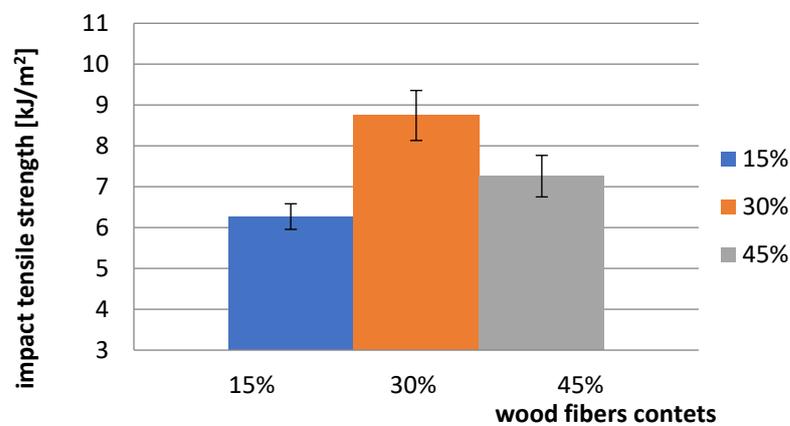


Figure 15. Impact tensile strength depending on the percentage of wood fibers for the PHBV–wood fiber composite.

It can be concluded that the PHBV–wood fiber composite with 30% wood fibers by weight had the highest impact strength.

**Temperature dimensional stability test**

The influence of temperature on the probe dimensions is shown in Figures 16–18.

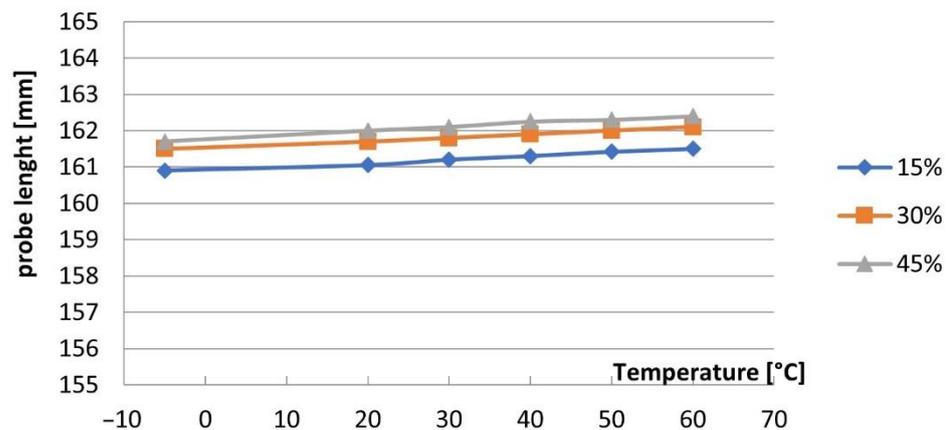


Figure 16. Influence of temperature on probe length.

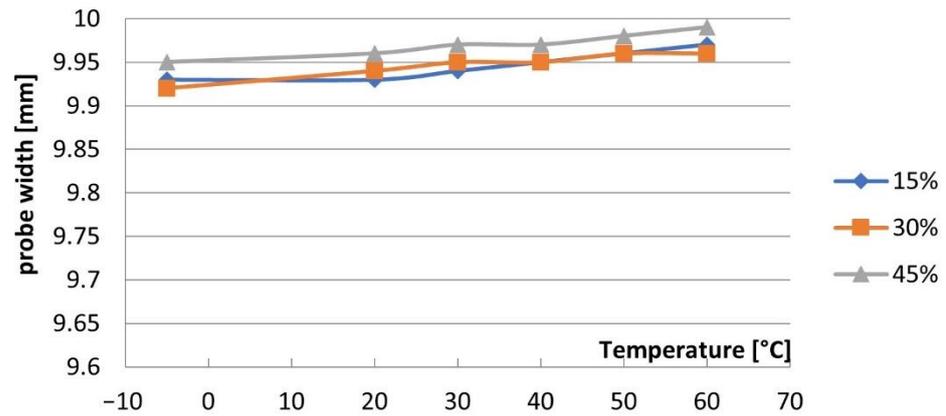


Figure 17. Influence of temperature on probe width.

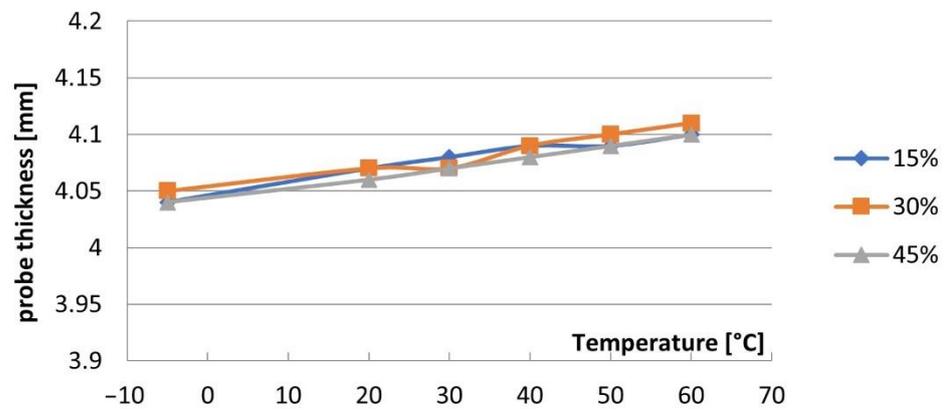


Figure 18. Influence of temperature on the thickness of probes.

**The dimensional stability test depending on water absorption**

It was found that the composite with 45% wood fibers was characterized by the highest water absorption. The largest increase in mass and an increase in the dimensions of the samples, in comparison with other samples, were observed. The lowest water absorption was observed for the WPC composite with 15% wood filler by weight. Its structure and color did not change significantly. The changes in water absorption for individual composites are shown in the diagrams (Figures 19–22).

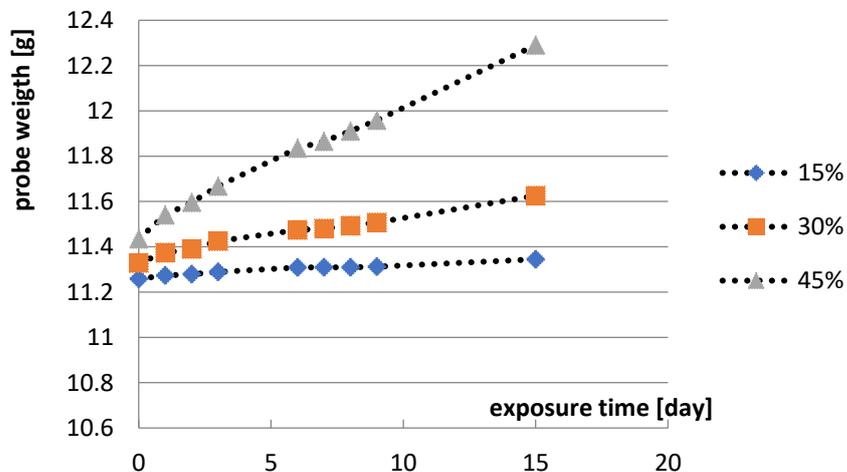


Figure 19. Weight of the probes from the PHBV-fiber composites with different wood fiber contents depending on the exposure time in water.

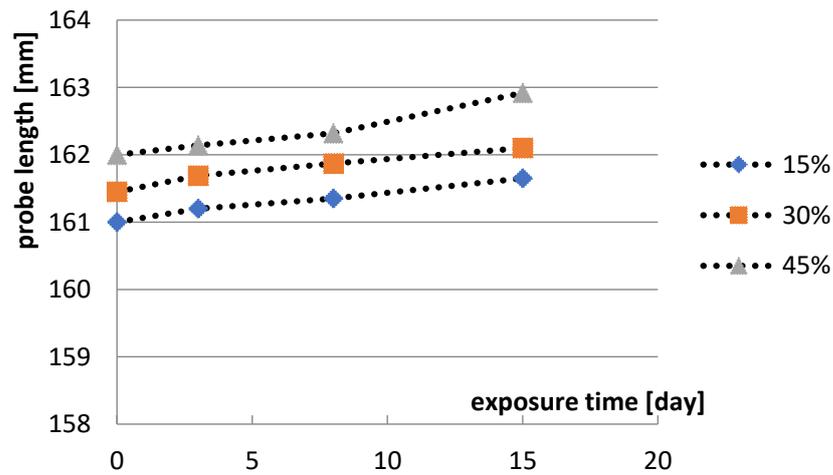


Figure 20. Change in the length of the PHBV–wood fiber composite probes under the influence of water absorption.

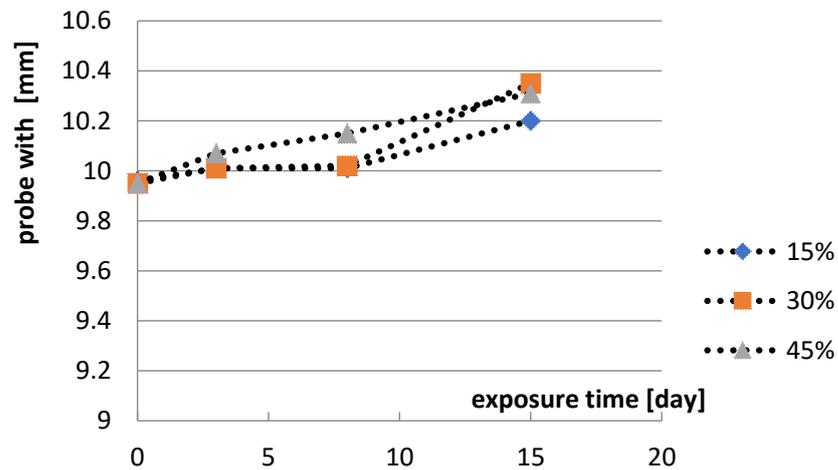


Figure 21. Changes in the width of the PHBV–wood fiber composite probes under the influence of water absorption.

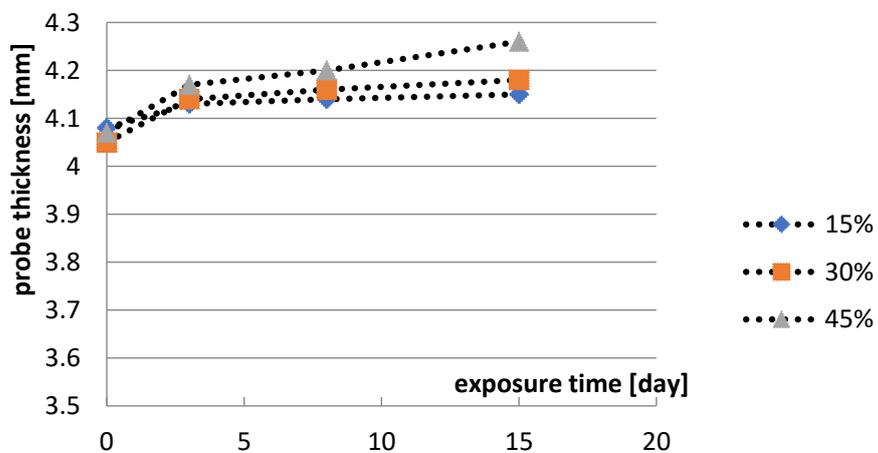


Figure 22. Changes in the thickness of the PHBV–wood fiber composite probes under the influence of water absorption.

After 2 days, a change in the color of the samples was observed. The least visible color changes were found in the WPC composite with the lowest content of wood fibers

by weight, i.e., 15%. A slippery coating was also observed on the surface of the samples during exposure to water. In addition, the weight and dimensions of the samples changed.

#### 4. The Results Analysis and Discussion

The results obtained bring closer the problems of manufacturing and using products made of WPC composites with a PHBV matrix and a filler in the form of wood fiber.

The increase in the content of wood fibers in the WPC composite caused a decrease in the primary longitudinal shrinkage from 1.02% (for 15% mas. Wood filler) to 0.93% and the secondary shrinkage from 1.15% to 1.03% for 45% wood fiber. The primary volumetric shrinkage also decreased from 1.04% to 0.76%, and the secondary shrinkage from 1.05% to 0.72%. The value of the primary transverse shrinkage increased from 2.13% to 2.64% with the increase in the wood fiber content from 15% to 45% by weight. The secondary transverse shrinkage also showed an increase from 2.33% to 2.82%. In the case of thickness shrinkage, both for the primary and secondary shrinkages, the highest value was shown in the PHBV composite with 30% wood fibers by weight. As the temperature increased, the dimensions of all samples made of all types of the produced and tested WPC composites increased. The largest change in dimensions was characterized by the WPC composite with 45% wood fibers by weight (increase of 0.43%). Analyzing the obtained results, it can be concluded that the content of natural fibers in the polymer may significantly affect various properties of the product, including changes in shrinkage during the production process. Natural fibers can reduce the overall shrinkage of a product by reducing internal stresses. Adding natural fibers can reduce shrinkage, especially if the fibers are evenly dispersed and well-oriented in the material. An increase in the amount of fibers in the polymer material can lead to an increase in resistance to longitudinal deformation. A larger amount of fibers can counteract shrinkage, creating a stronger and more stable internal structure of the product [39–41].

Samples with 30% wood fibers by weight exhibited the highest hardness in the area near the gate. The hardness of the PHBV–wood fiber composites with 15% and 45% filler by weight oscillated around 100 N/mm<sup>2</sup>. The hardness tested in the area opposite the gate increased with the distance from the end edge of the sample. The samples with 30% wood fibers by weight were characterized by the highest hardness. The hardness measured at the same point for different samples had the lowest value for this composite with 15% and 45% wood fibers by weight. Increasing the content of natural fibers in the polymer may have various effects on the Brinell hardness, which is a measure of a material's resistance to the penetration of a foreign body into its surface. An increase in the amount of natural fibers in the polymer can increase the Brinell hardness. The fibers provide reinforcement for the polymer structure, which can prevent the penetration of foreign bodies, leading to increased surface hardness [42]. In addition, the issue of uniformity and quality of fibers is very important. Fibers with a uniform length and thickness, without damage or impurities, are more effective in improving hardness. Inhomogeneities in the fiber structure may lead to local changes in hardness [43].

With the increase in the wood fiber content of the tested WPC composite, based on the tests in the static tensile test, the strains decreased (from 1.19% for 15% to 1.009% for 45% filler content by weight) and the tensile strength decreased (from 31.7 MPa (for 15%) to 27.8 MPa for 45% of filler content by weight). The fractures of the samples after stretching were evaluated organoleptically. With the increase in wood fibers, the homogeneity content of WPC decreased, the fibers were more visible, and the color of the samples became darker. For the composite with 30% wood fibers, the segmentation of the filler was visible. The highest impact tensile strength was observed for samples with a mass content of 30% wood fibers. The samples with 15% filler content showed the lowest impact tensile strength. Increased tensile strength was one of the key effects of increasing the natural fiber content in the polymer. Longer fibers with higher density and an even distribution in the polymer structure can significantly increase a composite's resistance to tensile forces. These fibers create a kind of scaffolding that prevents deformations, leading to an increase in the strength

of the material [44,45]. As the natural fiber content increases, the stiffness of the composite also increases. Young's modulus, which measures the stiffness of a material, increases, meaning that the composite is more resistant to deformation under load. This feature is particularly important in applications where high stiffness and resistance to deformation are required. However, increasing the natural fiber content also has its limitations. As the amount of fibers increases, the composite becomes less plastic. This means that it has a lower ability to plastically deform under load, which may be a disadvantage in some applications where the flexibility of the material is crucial [46,47]. Furthermore, increasing the fiber content may lead to the greater brittleness of the material. Despite increased tensile strength, composites with a higher fiber content may be more susceptible to cracks and fractures when overloaded. This brittleness results from a limited ability to plastically deform and may be problematic in applications requiring crack resistance. The increased content of natural fibers may also affect the elongation to break of the material. Materials with a higher fiber content are usually less flexible and more brittle. This means that the elongation at break can be limited, which is important for materials that need to have some elasticity to avoid cracking when loaded [48,49].

With the increase in the wood fibers share in the PHBV–wood fiber composite, the water absorption during the test of the absorption of samples increased. The largest weight gain (7.4%) took place for the composite with 45% wood fibers by weight. The weight changed the least in the case of the analyzed composite with 15% content of the filler (increase of 0.76%). All received samples of the WPC composite with the PHBV matrix were swollen in water. The largest increase in linear dimensions was observed for the WPC composite with 45% wood fiber content by weight (max. increase of 0.56%). The smallest dimension changes could be observed in the case of the WPC composite with a mass content of 15% wood fibers by weight. Based on the results obtained, it can be concluded that changing the content of natural fibers in the polymer significantly increased the water absorption of the composite. The key factor is the porosity of the composite structure. Natural fibers, especially longer ones, create intracellular spaces that are potential places for water retention. The more fibers, the greater the amount of pore spaces, which increases the ability of the composite to absorb water [50,51]. Additionally, some types of natural fibers, such as cellulose fibers, have a natural ability to absorb water. Introducing more such fibers into the composite structure increases its water absorption. The orientation of the fibers in the structure also plays a role—fibers arranged parallel to the composite surface can create more expensive paths for water, which further increases absorbency [52,53]. It is also worth paying attention to the impact of chemical modifications of natural fibers. Processes such as oxidation or other surface modifications can change the water absorption properties of fibers, which consequently affects the absorbency of the composite. Appropriate management of these factors is crucial for the design of composites with specific water absorption. In applications where moisture is a problem, it is necessary to select appropriate fiber proportions, their orientation, and possible chemical modifications to control the water absorption of the composite [54–56].

The research undertaken in this work is a continuation of research conducted by various scientists on WPC composites. An important problem in the production of composites of this type is the thermal degradation temperature of the fillers used in production: fibers or wood flour. This temperature allows the use of only those polymers that have a relatively low (approx. 160 °C) processing temperature. In the presented works, the trend of using mainly synthetic polymers is noticed. Nevertheless, some work was also carried out using the PHBV biopolymer. Unfortunately, the costs of purchasing such materials are relatively high. Various fillers were used in the conducted works and various properties were tested.

In terms of conducting research on PHBV composites with wood fillers, the literature does not directly refer to the use of the Arbocel C350 SR filler in the production of WPC. The existing literature concerns the production of PHBV biocomposites with selected fillers of a natural origin. One can find publications that also include wood fillers in general.

However, the research undertaken in them does not fully relate to the research presented in this paper.

For example, in the work [57], biocomposites with a PHBV matrix containing 10–40% maple wood fibers by weight were produced. The influence of increasing the share of wood fibers on the mechanical, thermomechanical, and morphological properties of the bio-composites was assessed. The tensile and flexural modulus of biocomposites with 40% mass fraction was improved by approximately 167% compared with that of pure PHBV. The theoretical values of the tensile modulus of elasticity of PHBV-based biocomposites, obtained from the Halpin–Tsai and Tsai–Pagano equations were consistent with the experimental values of the tensile modulus of elasticity. The heat deflection temperature was increased by 21% and the coefficient of linear thermal expansion decreased by 18% for the same amount of filler.

On the other hand, in publication [58], a biocomposite of PHBV–wood flour from oak wood was produced in the extrusion process, where the samples were successively injected. Among other things, processing and silanization possibilities, as well as the action of maleic anhydride on the filler, and consequently the composite, and the properties in the uniaxial tensile test were evaluated. The addition of 46% vol. of silanized filler allowed improving the modulus of elasticity by approx. 165% compared with pure PHBV. An improvement in the Young's modulus of 170% was also noted for the composite containing 28 vol. wood flour modified with maleic anhydride. The addition of a modified and pure filler decreased the degree of crystallinity of the matrix phase and resulted in an increase in the brittleness of the composites, which brought in a reduction in the tensile stresses and deformation of the samples.

In the publication by Vandi and co-authors [59], the commercial feasibility of extruding the PHBV–wood filler biocomposite, even at temperatures as high as 190 °C, was investigated by optimizing the screw speed and feed rate, and consequently the induced shear rate and residence time. In addition, it was found that the mechanical properties of the PHBV composite with 40% wt. wood were better than those described in the literature. Compared with pure PHBV, a 73% increase in the Young's modulus and 80% retention of tensile strength was obtained. Taguchi orthogonal plans were adopted for the study design and optimization in order to examine the effect of extrusion parameters (temperature profile, screw speed, dosing speed, and fiber mixing) on the processing of pure PHBV biopolymer and wood–PHBV composites with wood contents of 10, 20, 30 and 40 wt.%. The evaluation of the mechanical properties was carried out by testing the tensile strength, tensile modulus, and strain at maximum tensile stress. It was found that, both in the case of pure PHBV and the biocomposite, the molecular weight decreased under the influence of high shear stress and long residence time in the plasticizing system from 550–650 kDa to 350–550 kDa. However, the decrease in molecular weight was not large enough to cause a significant reduction in the mechanical properties of the products. According to the authors, the results of these studies are of great importance in the production of biocomposite products in the industrial dimension because the processing window is highly narrow and thermal degradation can be limited by optimizing the adjustable parameters of the extrusion process.

In the work by Singh et al. [60], a biocomposite based on PHBV with wood fiber and talc was produced in the extrusion process, and the samples were then injected. A clear improvement of 200% in the Young's modulus and double-strength bending was shown, i.e., 20% wt. talc and 20% wt. wood fiber in the PHBV matrix. The dynamic mechanical and thermomechanical properties were tested experimentally and a similar trend was shown. In addition, a quantitative decrease in the coefficient of linear thermal expansion of 36% and an improvement in the heat deflection temperature of the hybrid composite were noted.

In publication [61], an analysis of the micro and macroscopic changes that occur during the biodegradation of poly(3-hydroxybutyrate-co-3-hydroxyvalerate) (PHBV) composites and wood flour in soil was carried out. Composites containing various filler contents, i.e., 0, 20%, and 50% wt. were buried in the soil at a research site in subtropical Australia,

where the results were compared with those for polylactic acid (PLA) and polyethylene (PE) based on wood composites as a benchmark. It was noted that the degree of degradation of the PHBV–wood composites increased with the content of wood filler, with a mass loss after 12 months five times greater for PHBV with 50% filler content compared with the pure PHBV samples tested under the same conditions. The pure PHBVs retained their mechanical properties after 12 months underground. The PHBV composite with 50% filler content lost its mechanical properties after the same period with a 13% weight loss. The PLA and PE matrix composites recorded only minimal decreases in mechanical properties, which can be attributed to the degradation caused by increased humidity/loosening only on the surface.

In reference [62], a biocomposite was produced from the PHBV/PBAT blend (weight ratio 70:30) with a share of recycled wood fibers of 10% by weight and 2 wt. nanoclay content, where the composite was injected both conventionally and using the microcellular method. It was noted that the addition of 10% wood fibers decreased the pore size and increased the density of the finished product, increasing the Young's modulus and tensile strength compared with the pure porous blend. In addition, unlike the pure PHBV/PBAT blend, the microcellular composites showed higher tensile strength and strain at break compared with their solid counterparts. The degree of crystallinity of PHBV increased significantly in both the solid and microcellular composites, although the degree of crystallinity of PHBV in the solid components was slightly higher than that of their microcellular counterparts. In addition, it was noted that the addition of 2% nanoclay did not cause significant changes in the cell morphology and mechanical properties of the composite, but improved the thermal stability.

Research by Chan and co-authors [63] aimed at evaluating the properties of the material, especially long-term stability in rooms for a biocomposite based on pine wood flour and poly(3-hydroxybutyrate-co-3-hydroxyvalerate) (PHBV) modified with boron nitride and an additional filler in talc form. Both boron nitride at 1% wt. and talc at 5% wt. had an effect on the nucleation and crystallization of the polymer, with boron nitride being more effective. When 5 and 10% by weight of talc were added, the tensile strength of the wood-based PHBV composite (50% by weight) increased from about 22.3 MPa to about 24.3 MPa and 24.7 MPa, respectively. The tensile modulus increased from about 5.6 GPa to about 6.9 GPa and 8.1 GPa, respectively. It was observed that talc could fill voids in the composite matrix and/or serve as an alternative filler by modifying the deformation mechanism of the composite. It was also noted that the modulus decreased after one year of aging under controlled conditions, i.e., at 20 °C and 50% humidity. It was suggested that the interaction of wood swelling by moisture absorption, shrinkage of the PHBV due to secondary crystallization, and local tension around the talc particles led to a weakening of the adhesive bond between the polymer matrix and the talc.

In the work by Chan et al. [64], the effect of three methods of improving compatibility on the mechanical properties of PHBV–wood fiber composites was examined. Silanization, maleic anhydride, and polymethylenediphenyl diisocyanate (pMDI) were used in the tests. The results showed that the use of silanization reduced the tensile strength of the composite by approx. 20%. The use of maleic anhydride, regardless of the amount, led to a slight increase in tensile strength (approx. 5%) and impact strength (approx. 10%). The addition of 2 wt. polymethylene diphenyl diisocyanate increased the tensile strength by about 25%, but after adding 4% by mass, the strength returned to the initial value. The method of manufacture, i.e., extrusion, had a significant impact on the properties of the composite, causing a decrease in tensile strength by approximately 40% and tensile strength by approximately 45% when extrusion with higher shear was used.

## 5. Conclusions

The production of WPC composites is still a current trend in the production of plastics that enables the use of, for example, post-production waste (wood processing waste).

The list of polymeric materials that can be used for this purpose is short and processing problems still occur.

In this study, the dimensional stability, shrinkage, and selected mechanical, processing, and functional properties of WPC composites of the wood fiber–PHBV with different contents of wood fiber were examined. The PHBV matrix can be successfully used for the production of such materials. The following conclusions can be drawn from the conducted research.

In the range of the tested amounts of filler, i.e., 15–45% in the PHBV–wood fiber composite, an increase in the percentage of wood fibers caused a decrease in primary shrinkage. With the change in the content of wood fibers in the PHBV matrix, the weight of the molded pieces changed. Large differences in the weights of the samples produced in the series were found in the WPC composite with a mass content of 45% wood fibers. With a large amount of fibers, there were problems in filling the mold cavities.

The hardness of the samples tested near the gate showed no tendency to change. The samples with a mass content of 30% wood fibers were characterized by the highest hardness. The hardness of WPC composites with mass contents of 15% and 45% wood fibers oscillated around 100 [N/mm<sup>2</sup>] in the Brinell method. The hardness tested in the area opposite the gate showed an increase with the distance from the end edge of the sample. The samples with a mass content of 30% wood fibers were also characterized by the highest hardness. The hardness values measured at the same point for different samples had the smallest values for the WPC composite with 15% and 45% by weight of wood fibers.

Tensile strength is satisfactory and decreased by a maximum of about 15% with the increase in the mass content of wood fiber in the tested WPC composite. The highest impact tensile strength was shown by samples with a mass content of 30% wood fibers.

As the temperature increased (−5 ÷ 60 °C), the dimensions of the samples made of all types of the produced WPC composite increased by a maximum of 1.5%. The largest change in dimensions was characterized by the WPC composite with 45% wood fibers mass content.

With the increase in the mass increase of wood fibers in the WPC composite, the amount of water absorbed during the test increased. The largest weight gain occurred for the WPC composite with a mass content of 45% wood fibers. The weight changed the least in the case of the WPC composite with a mass content of 15% of the filler.

Processing problems are related to both the shrinkage of the material and the occurrence of the Barus effect during the extrusion of the composition, depending on the amount of wood fiber.

The properties of the tested types of biocomposite allowed for the assumption that they can be used in the production of a wide range of various WPC products. Reducing the price of the product compared with the use of pure PHBV was significant in the case of using 45% of the filler. Future work in this field should focus on assessing the possibility of using the PHBV–wood fiber biocomposite in specific WPC applications and ways to improve the properties of this type of biocomposites in terms of flexibility and reducing brittleness.

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