Influence of a bark-filler on the properties of PLA biocomposites

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ABSTRACT

In this study, wood plastic composites (WPC) made of poly(lactic acid) PLA and a bark-filler were manufactured. Two degrees of bark comminution (10–35 mesh and over 35 mesh) and varied content of bark (40, 50 and 60%) were investigated. The studied panels were compared with analogically manufactured HDPE boards. The manufacture of composites involved two stages: at first, WPC granules with the appropriate formulation were produced using the extruder (temperatures in individual extruder sections were 170–180 °C) and crushing using a hammer mill after cooling the extruded composite; secondly, the obtained granulate was used to produce boards with nominal dimensions of 300 × 300 × 2.5 mm3 by flat pressing in a mold, using a single daylight press at a temperature 200 °C. The study proved that comminuted bark can be applied as a filler in PLA composites. However, an increase in bark content decreased mechanical properties (MOR, MOE) and deteriorated humidity resistance (high TS and WA) of the panels. Along with the increase in bark content, an increase in the contact angle of the composite surfaces and a decrease in the total surface energy were noted. It was also found that PLA composites have higher strength parameters and lower moisture resistance compared to HDPE composites with the same bark content.
Introduction

The dynamic development of wood plastic composites (WPC) technology involves the introduction of new material solutions for the matrix and the filler. In both cases, the biodegradability of raw materials is essential. Nowadays a significant amount of WPC composites known as biocomposites is produced with the addition of polymers derived from fossil fuels. These include primarily polyethylene PE (used in decking and construction, consumer goods), polypropene PP (used in automotive, construction, consumer goods) or polyvinyl chloride PVC (used in decking and construction) [1]. However, these materials are not biodegradable; thereby, it makes it debatable whether these WPC composites should be included in the family of biocomposite materials.

Biodegradable thermoplastics such as poly(lactic acid) (PLA) or polyhydroxyalkanoate (PHA) can be used in the production of WPC composites. PHA slowly decomposes into water and carbon dioxide under activity of microorganisms present in soil, sewage or silt; anaerobic conditions are particularly favorable for this process. That makes PHA applicable in packaging and components with a short shelf life. In turn, PLA is less biodegrade under natural conditions; therefore, it can be applied in components with a long service life. Environmentally safe degradation of PLA occurs during composting [2]. PLA is by far the most widely studied and applied aliphatic polyester in history. Regarding its advantages it is a leading biomaterial for multiple applications in medicine and in the industry replacing conventional petrochemical polymers [3]. However, its sensitivity to moisture, susceptibility to aging, limited impact strength and high rigidity in some applications [4] result in frequent modification of PLA. One of the modifications involves introduction of fillers (manufacturing WPC composite), especially in construction applications.

The most popular natural fillers are wood materials in the form of wood flour or wood fibers. The size of filler’s particles depends on the purpose of the composite [5]. In general, there are no limitations to the use of different types of wood as fillers for polymer composites [6]. However, due to the simpler anatomical structure and higher ratio of length to diameter of the elements of anatomical structure, it is preferable to use coniferous species [7]. Saputra et al. [8] and Shebani et al. [9] reported a negative impact of extractive substances content on bonding wood fibers with a polymer matrix and on the thermal stability of composites. Aside solid wood, WPC composites production can also apply sawdust and flour, post-use wood materials such as particleboards and MDF [10] or waste paper [11, 12]. Particles obtained from plant materials other than wood can be also utilized as fillers for WPC composites.

Bark is a common waste in the production of wood and wood materials. The main two trends in bark applications are: bark obtained in a special way used in the production of medicines, cosmetics, tannins, dyes, cork [13, 14] and waste bark for composting or
incineration [13, 15]. The efficiency of the incineration process is low due to the typical for bark high humidity (34–40% during dry summer, and up to 60% in winter) and a high content of mineral substances constituting ash after burning (more than 2%) compared to wood [13]. Bark can be utilized as an alternative raw material for the production of particleboard [15, 16]. The tests generally showed a decrease in panel strength parameters along with an increase in the bark share. Shredded bark is also a potential filler in the production of WPC composites [17–20]. Yemele et al. [18] showed that WPC composites filled with bark have lower strength parameters compared to analogous composites filled with wood flour. Also, other researchers reported a decline in the mechanical properties of WPC composites filled with bark [17, 20]. In turn, the use of bark as a filler in WPC composites improves their physical properties related to moisture absorption. Gozdecki et al. [19] and Najafi et al. [21] indicated that the increase in bark content in WPC composites decreases their swelling and water absorption.

Natural fillers have also been used in research on biodegradable WPC composites applying PLA as a polymer matrix. Among them were: wood fibers [22–24], wood flour (WF) [25, 26], cellulose nanofibrils [27], cork [25, 28], bamboo fiber [29], abaca fibers [30], rubber wood sawdust [31], hemp fibers [32]. In general, authors report an improvement in the mechanical properties of PLA composites filled with wood fibers with a filler content of up to 20% [23] or 30% [22]. Csizmadia et al. [33] showed a beneficial effect of wood fiber modification with phenol–formaldehyde resin on improving strength properties and reducing water absorption in the production of PLA-based composites. In turn, Andrzejewski et al. [25] showed the beneficial effect of cork filler (up to 30%) on the dimensional stability of PLA composites in the presence of moisture. However, the use of wood flour as a filler affects resistance of PLA composites to moisture [26].

As part of this study, the effect of the addition of bark as a filler to the PLA matrix in the production of biodegradable WPC composite was determined. This type of composite variant has not yet been studied. For comparison, an analogous WPC composites based on high-density polyethylene (HDPE) matrix were made. Three levels of bark content at two degrees of bark comminution were investigated. The manufacture of the composites involved two stages: extrusion and flat pressing. The effect of bark addition was analyzed in the context of selected physical and mechanical properties of composites.

**Experimental**

Twelve variants of WPC composite panels (Table 1) were manufactured based on two types of polymer matrices: polylactic acid—PLA (Ingeo™ Biopolymer 2003D, NatureWorks LLC, Minnetonka, MN, USA), and high-density polyethylene—HDPE (Hostalen GD 7255, Basell Orlen Polyolefins Co., Plock, Poland). The filler was a crushed pine bark supplied from a sawmill. The bark was dried to a moisture content of 5% and then mechanically ground and sorted into two variants:

1. Particles passing through a 2-mm sieve (approx. 10 mesh) and remaining on a 0.49-mm sieve (approx. 35 mesh)—Fig. 1a.
2. Particles passing through a 0.49-mm sieve (over 35 mesh)—Fig. 1b.

No other additives commonly used in the production of WPC such as compatibilizers were applied in the study, due to comparative assessment of combinations: PLA—bark and HDPE—bark.

The manufacture of composites involved two stages. At first, WPC granules with the appropriate formulation (Table 1) were produced using the Leistritz Extrusionstechnik GmbH, Nürnberg, Germany, extruder (temperatures in individual extruder sections were 170–180 °C). The obtained continuous composite web was ground on a hammer mill. Secondly, the obtained granulate was used to produce boards with nominal dimensions of 300 × 300 × 2.5 mm³ by flat pressing in a mold, using a single daylight press at a temperature of 200 °C and a maximum unit pressing pressure \( p_{\text{max}} = 1.25 \text{ MPa} \) (pressure during pressing, along with plasticization of the material, was gradually increased from 0 to \( p_{\text{max}} \)). Pressing time was 6 min. After hot pressing, the plates were cooled in the mold for 6 min in a cold press (approx. temp. 20 °C). Eventually, the manufactured panels were conditioned at ambient temperature and humidity for 7 days under laboratory conditions (20 ± 2 °C, 65 ± 5% humidity).

The following physical and mechanical properties of the boards were tested:
Density according to EN 323:1999 and density profile using Laboratory Density Analyser DAX GreCon (Fagus-Grecon Greten GmbH & Co. KG, Alfeld, Germany). Density measurement was made every 0.02 mm at the measurement speed of 0.05 mm/s.

Modulus of rupture (MOR) and modulus of elasticity (MOE) according to EN 310:1994 [34].

Screw holding (SH) according to EN 320:2011 [35].

Thickness swelling (TS) and water absorption (WA) after 2-h and 24-h immersion in water—according to EN 317:1999 [36].

Wettability (contact angle) and surface free energy. Contact angle (θ) based on the sessile drop method and performed on a Phoenix 300 (Surface Electro Optics, Suwon City, Korea) contact angle analyzer, equipped with microscopic lenses and digital camera was measured. The distilled water and diiodomethane were used as reference liquids for wettability calculations. The values of contact angles were determined after 45 s of application of drops of liquid onto the surface of the reference (the water). For each type of board (on both sides) ten droplets were measured.

The surface free energy was determined using the Owens–Wendt (1969) method.

Tests involved ten replicates of each variant. Additionally, the microscopic images of samples using a Tabletop Microscope TM3000 microscope (Hitachi Ltd., Tokyo, Japan) were taken.

Statistical analysis of the results was carried out using Statistica version 13 (TIBCO Software Inc., CA, USA). The analysis of variance (ANOVA) was used to test \( \alpha = 0.05 \) for significant differences between factors. A comparison of the means was made using Tukey’s test, with \( \alpha = 0.05 \). In addition, principal component analysis (PCA) was performed to describe the patterns of covariant among the examined traits.

### Table 1

Characteristics of the composition of individual variants of composites

| Variant | Matrix | Share of matrix | Bark share |
|---------|--------|-----------------|------------|
|         |        |                 | Small particles over 35 mesh | Large particles 10–35 mesh |
| I       | PLA    | 60              | –          | 40          |
| II      | PLA    | 50              | –          | 50          |
| III     | PLA    | 40              | –          | 60          |
| IV      | PLA    | 60              | 40         | –          |
| V       | PLA    | 50              | 50         | –          |
| VI      | PLA    | 40              | 60         | –          |
| VII     | HDPE   | 60              | –          | 40          |
| VIII    | HDPE   | 50              | –          | 50          |
| IX      | HDPE   | 40              | –          | 60          |
| X       | HDPE   | 60              | 40         | –          |
| XI      | HDPE   | 50              | 50         | –          |
| XII     | HDPE   | 40              | 60         | –          |

Figure 1: Particles of bar:

a Large particles (10–35 mesh).

b Small particles (over 35 mesh).
Results and discussion

The tested panels were characterized by densities ranging between 1114 and 1182 kg/m³ for PLA matrix and 1051 and 1105 kg/m³ for HDPE. The average density values for individual panel variants are presented in Table 2. The density variation for individual variants within the same matrices (PLA or HDPE) did not exceed 6%. Also, no effect of filler’s particle size was observed. Moreover, the PLA boards consisting the same share and size of filler particles had higher density by 1 to 11%, respectively. This is due to the higher density of PLA matrix compared to HDPE matrix. Similar dependencies were observed by Andrzejewski et al. (2019) [25] studying WPC composites based on PLA and PP. The authors also pointed to a decrease in the density of PLA-based composites with an increase in cork content. A similar phenomenon was recorded in the present study for bark content of 40–50%. It should be added, however, that a further increase in the bark content (up to 60%) would increase the density of the composite. All variants of the tested panels were characterized by an even distribution of density across the cross section (Fig. 2). Density differences across the thickness of individual panels generally did not exceed 100 kg/m³. This demonstrates good homogenization of composite components and even distribution of filler particles in the polymer matrix.

The strength parameters of the composites presented in Table 3 were higher for PLA panels compared to the HDPE panels. This is due to the greater rigidity of PLA than polyolefins [37].

The increase in bark content regardless of its size resulted in a significant decrease in the MOR value. Similar relationships demonstrated Gozdecki et al., Yemele et al. or Andrzejewski et al. [18, 19, 25], where the authors while studying composites with a lower bark content (max. up to 40%) recorded a nearly sevenfold decline in MOR for PLA composites with small particles of bark, and the maximum decrease was about 60%. This relationship was not found in the present study for HDPE composites. The addition of bark to HDPE regardless the size of particles resulted in a comparable decrease in strength, maximum 30%.

The increase in bark content from 40 to 60% significantly affected the decrease in MOE values only for PLA composite with the addition of small particles of bark. At the same time, higher MOE values were recorded for PLA matrix composites with large particles of bark compared to small particles. An opposite effect was obtained for HDPE matrix composites which showed significantly higher values for small particle. These results are borne out by two other studies: Yemele et al. and Çetina et al. [18, 38].

The increase in the content of bark particles in the tested composites also influenced the decrease in SH value, and it varied depending on the particles size. Similarly as in the case of MOE, the largest decrease was recorded for PLA matrix composites with the addition of small particles of bark. Gozdecki and Kociszewski [39] when studying PP-based composites observed a decrease in SH value with an increase in wood flour content. In turn, Falk et al. [40] found that for composites based on LDPE and PP and wood flour the increase in lignocellulosic filler content did not affect SH values.

When analyzing the strength properties, it must be emphasized that small particles of bark have about 20 times larger specific surface area in comparison with large particles, which significantly increases the potential contact surface between the filler and the matrix, as well as increases their dispersion in the

Table 2 The density of manufactured panels according to variants

| Matrix | Bark share [%] | Large particles of bark | Small particles of bark |
|--------|----------------|-------------------------|-------------------------|
|        |                | Average density [kg/m³] | Standard dev. [kg/m³]   | Average density [kg/m³] | Standard dev. [kg/m³] |
| PLA    | 40             | 1167                    | 47                      | 1171                    | 41                      |
| PLA    | 50             | 1159                    | 65                      | 1114                    | 46                      |
| PLA    | 60             | 1182                    | 58                      | 1123                    | 51                      |
| HDPE   | 40             | 1051                    | 24                      | 1053                    | 26                      |
| HDPE   | 50             | 1078                    | 25                      | 1105                    | 27                      |
| HDPE   | 60             | 1094                    | 29                      | 1077                    | 38                      |
matrix. At the same time, the matrix structure is thinner, which with a rigid but brittle PLA material can also affect the strength of composites. Faludi et al. [41] reported that the decrease in strength of composites with the addition of lignocellulosic fillers on one hand may be due to limited interfacial adhesion, which depends on the difference in surface energy of the combined components, and on the other hand it is the effect of cracks occurring in the filler itself. The tested PLA boards were characterized by free surface energy in the range from 51.676 to 37.755 mJ m\(^{-2}\), while the value decreased with the increase in the content and degree of bark fragmentation (Table 4). There are no data in the literature regarding the surface energy of clean bark. However, taking into account the results obtained for the tested panels and similar literature data on the surface energy of PLA and wood [42, 43], it can be assumed that the decrease in composites strength noted in the present

Figure 2  Density profiles of manufactured panels.

Table 3 Mechanical properties of manufactured boards

| Share of bark [%] | PLA          | Average       | HDPE          | Average       |
|-------------------|--------------|---------------|---------------|---------------|
|                   | L   | S   |          | L   | S   |          | L   | S   |          |
| MOR [N/mm\(^2\)] |     |     |          |     |     |          |     |     |          |
| 40                | 33.19\(^{ab}\) | 34.21\(^{a}\) | 33.7\(^{A}\) | 20.08\(^{de}\) | 20.83\(^{d}\) | 20.46\(^{CD}\) |
| 50                | 28.24\(^{c}\) | 16.67\(^{ef}\) | 22.45\(^{BC}\) | 19.03\(^{de}\) | 18.21\(^{de}\) | 18.62\(^{D}\) |
| 60                | 30.62\(^{bc}\) | 14.27\(^{e}\) | 23.35\(^{B}\) | 14.31\(^{e}\) | 16.86\(^{ef}\) | 15.58\(^{E}\) |
| Average           | 30.68\(^{1}\) | 22.25\(^{2}\) | –             | 17.81\(^{3}\) | 18.63\(^{3}\) | –             |
| MOE [N/mm\(^2\)] |     |     |          |     |     |          |     |     |          |
| 40                | 3597\(^{a}\) | 3669\(^{a}\) | 3634\(^{A}\) | 1463\(^{f}\) | 1854\(^{de}\) | 1659\(^{C}\) |
| 50                | 4049\(^{b}\) | 2990\(^{b}\) | 3520\(^{AB}\) | 1771\(^{de}\) | 1895\(^{cd}\) | 1836\(^{C}\) |
| 60                | 3944\(^{a}\) | 2345\(^{c}\) | 3233\(^{B}\) | 1595\(^{de}\) | 2035\(^{ed}\) | 1827\(^{C}\) |
| Average           | 3864\(^{1}\) | 3048\(^{2}\) | –             | 1605\(^{4}\) | 1928\(^{3}\) | –             |
| SH [N]            |     |     |          |     |     |          |     |     |          |
| 40                | 219.55\(^{a}\) | 228.36\(^{a}\) | 224.19\(^{A}\) | 151.72\(^{be}\) | 154.29\(^{be}\) | 153.00\(^{B}\) |
| 50                | 164.96\(^{b}\) | 99.68\(^{d}\) | 132.32\(^{C}\) | 145.71\(^{be}\) | 150.07\(^{bc}\) | 147.89\(^{B}\) |
| 60                | 215.35\(^{a}\) | 96.63\(^{d}\) | 159.11\(^{B}\) | 98.90\(^{d}\) | 136.55\(^{c}\) | 117.73\(^{C}\) |
| Average           | 199.28\(^{1}\) | 143.11\(^{2,3}\) | –             | 132.11\(^{3}\) | 146.97\(^{2}\) | –             |

L large particles of bark, S small particles of bark, A,B,…, a,b,…, 1,2,… homogenous group
study is mainly affected by susceptibility of bark for cracking.

Figure 3 shows photographs taken with the use of a scanning electron microscope, enabling observation of the topography of individual variants of the tested composites, in which, regardless of the share and size of the bark particles, cracks are visible in the lignocellulosic particles. The cracks are formed at several stages of the process, e.g., when material is conveyed by the screw or forced through the die in the extruder [44].

All the three variable factors, i.e., matrix type, filler particles size and filler share, had a significant effect on the MOR, MOE and SH values of the manufactured composites (Table 5). However, the percentage of contribution (P%) for individual factors as well as for their interactions was varied.

The most significant factor that affected MOR values was the type of matrix (P = 29.36%), followed by the share of bark (P = 22.65%) and the size of bark particles (P = 7.64%). The total percentage of contribution for these factors was 59.65%, which proves their importance. A significant interaction was
demonstrated between the matrix type and the bark particle size (11.05%). Interestingly, the influence of filler particle size on MOR was less than the effect of factors not considered in this study (error = 9.27%).

Type of matrix was found to have the highest percentage share (68.17%) of influence on MOE values. Interestingly, the share of bark filler indicated no significant effect on the parameter. The effect of filler particles size on the MOE values and the effect of interaction between the share of bark-filler and the filler particles size were less than the effect of factors not considered in this study (error = 12.77%).

When analyzing the impact of individual factors on SH values, all sources of variance were significant. In contrast to MOR and MOE, the most crucial factor in determining the SH value was the share of bark-filler (P = 25.29%), followed by the interactions between matrix type and size of bark particles (P = 15.30%) and interaction between matrix type and share of bark (P = 14.61%). The error value (P = 11.35%) was higher than the P values of the rest tested factors; this indicates that untested factors had a greater influence on SH.

The physical parameters of the manufactured composites are present in Table 6. The PLA-matrix panels were generally characterized by lower water resistance (TS and WA) compared to HDPE-matrix panels. The difference was particularly noticeable after immersion for 24 h in the water, regardless of the bark content and the filler particle size. Moisture penetration into the internal structure of the composite occurs through lignocellulosic particles, which increase their dimensions during this process. This deteriorates the contact between the filler and the polymer mesh, and the effect being greater to stiffer PLA. The HDPE matrix with good flexibility can partially absorb changes in the dimensions of the filler. An increase in the filler’s compactness as well as an increase in its particle size results in a significant increase in TS and WA values. This is due to the greater availability of lignocellulosic filler particles on the composite surface [45, 46]. It is also confirmed by the lower contact angles of the surface with water when larger filler particles are used (Table 4). It is noteworthy that bark contains hydrophobic suberin [47, 48] which can reduce the wettability of the surface.

The most significant influence on TS and WA of tested boards had type of matrix and the share of bark-filler (Table 7). The total percentage of the impact of these factors after 24 h of immersion in water was over 55%. The treatment revealed also that size of bark particles had a minor influence on TS and WA (P = 5.39% and P = 1.91%, respectively). It should be noted that especially during the TS test error values for 2 h and 24 h were considerably high, which may have been caused by other external factors excluded from the research. The greatest impact on TS and WA out of tested interactions had the interaction among the type of matrix and the filler particle size and the filler content.

PCA was performed for further data evaluation. On the charge distribution graph (Fig. 4), the points denoting Variants I and IV have positive coordinates on both axes p1 and p2. Thus, they are at a small distance from each other and the points indicating
the properties of MOR and SH. It can therefore be concluded that these variants not only have similar properties, but also have the highest MOR and SH values. It should be emphasized that both variants were made of PLA. Points marking variants II and III have positive coordinates on the p1 axis, and the distance between them is small, which shows that in the case of PLA-based composites, the bark share within 50–60% does not significantly affect strength properties. The size of the bark particles used to make the composite has a much greater impact in this case, as evidenced by the distance between the points denoting variants II and III as well as V and VI. In the case of points corresponding to variants of HDPE-based composites, it can be stated that they are characterized by relatively good strength properties, and the impact of bark share and the size of bark particles slightly differentiate the overall strength properties of these panels. This is evidenced by the fact that the points corresponding to the variants of

Table 5  ANOVA for selected factors affecting MOR, MOE and SH of manufactured composites

| Source of variance | MOR | MOE | SH |
|--------------------|-----|-----|----|
|                    | p   | P (%) | p | P (%) | p | P (%) |
| a                  | 0.000 | 29.36 | 0.000 | 68.17 | 0.000 | 11.11 |
| b                  | 0.000 | 7.64 | 0.000 | 1.82 | 0.000 | 5.40 |
| c                  | 0.000 | 22.65 | 0.058 | 0.72 | 0.000 | 25.29 |
| a × b              | 0.000 | 11.05 | 0.000 | 8.58 | 0.000 | 15.30 |
| a × c              | 0.000 | 7.23 | 0.001 | 1.70 | 0.000 | 14.61 |
| b × c              | 0.000 | 5.74 | 0.000 | 3.21 | 0.000 | 4.46 |
| a × b × c          | 0.000 | 7.06 | 0.000 | 3.02 | 0.000 | 12.47 |
| Error              | 9.27 | 12.77 | 11.35 |

a matrix type/composite, b size of bark particles, c share of bark-filler, p probability of error, P percentage of contribution

Table 6  Selected physical properties of manufactured boards

| Share of bark [%] | PLA | Average | PE | Average |
|-------------------|-----|---------|----|---------|
|                   | L   | S       | L  | S       |     |
| TS 2 h [%]        |     |         |    |         |     |
| 40                | 1.42<sup>bc</sup> | 0.82<sup>d</sup> | 1.12<sup>C</sup> | 1.37<sup>bc</sup> | 1.04<sup>cd</sup> | 1.21<sup>BC</sup> |
| 50                | 2.20<sup>a</sup> | 1.62<sup>b</sup> | 1.91<sup>A</sup> | 1.34<sup>bc</sup> | 1.42<sup>bc</sup> | 1.38<sup>B</sup> |
| 60                | 2.17<sup>a</sup> | 1.43<sup>bc</sup> | 1.80<sup>A</sup> | 2.29<sup>a</sup> | 1.49<sup>b</sup> | 1.89<sup>A</sup> |
| Average           | 1.91<sup>1</sup> | 1.28<sup>3</sup> |     | 1.67<sup>2</sup> | 1.32<sup>3</sup> |     |
| TS 24 h [%]       |     |         |    |         |     |
| 40                | 4.63<sup>abc</sup> | 2.98<sup>de</sup> | 3.80<sup>B</sup> | 2.12<sup>ef</sup> | 1.72<sup>f</sup> | 1.92<sup>C</sup> |
| 50                | 3.90<sup>cd</sup> | 4.26<sup>bc</sup> | 4.05<sup>B</sup> | 2.16<sup>ef</sup> | 2.47<sup>ef</sup> | 2.31<sup>C</sup> |
| 60                | 4.90<sup>abc</sup> | 5.19<sup>ab</sup> | 5.04<sup>A</sup> | 5.55<sup>a</sup> | 2.74<sup>e</sup> | 4.15<sup>B</sup> |
| Average           | 4.46<sup>1</sup> | 4.08<sup>1</sup> |     | 3.28<sup>2</sup> | 2.31<sup>3</sup> |     |
| WA 2 h [%]        |     |         |    |         |     |
| 40                | 0.96<sup>ef</sup> | 0.52<sup>g</sup> | 0.74<sup>B</sup> | 0.70<sup>efg</sup> | 0.64<sup>fg</sup> | 0.67<sup>D</sup> |
| 50                | 2.13<sup>b</sup> | 1.08<sup>de</sup> | 1.60<sup>B</sup> | 0.80<sup>efg</sup> | 0.83<sup>efg</sup> | 0.82<sup>D</sup> |
| 60                | 1.70<sup>c</sup> | 3.76<sup>a</sup> | 2.73<sup>A</sup> | 1.46<sup>cd</sup> | 0.82<sup>efg</sup> | 1.14<sup>C</sup> |
| Average           | 1.59<sup>1</sup> | 1.72<sup>3</sup> |     | 0.99<sup>2</sup> | 0.77<sup>3</sup> |     |
| WA 24 h [%]       |     |         |    |         |     |
| 40                | 4.50<sup>d</sup> | 2.75<sup>e</sup> | 3.63<sup>C</sup> | 1.71<sup>fg</sup> | 1.05<sup>g</sup> | 1.38<sup>E</sup> |
| 50                | 7.16<sup>bc</sup> | 4.23<sup>d</sup> | 5.70<sup>B</sup> | 2.25<sup>ef</sup> | 2.43<sup>ef</sup> | 2.34<sup>D</sup> |
| 60                | 6.39<sup>c</sup> | 11.24<sup>a</sup> | 8.81<sup>A</sup> | 7.44<sup>b</sup> | 2.88<sup>e</sup> | 5.16<sup>B</sup> |
| Average           | 6.01<sup>1</sup> | 5.90<sup>1</sup> |     | 3.80<sup>2</sup> | 2.12<sup>3</sup> |     |

L large particles of bark-filler, S small particles of bark-filler, A,B,..., a,b,..., 1,2,... homogenous groups

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HDPE-based composites are located in the same quarter of the graph, at short distances from each other. Only the point denoting variant 10 with poor strength properties is located at a considerable distance from the other points denoting variants of HDPE-based composites.

Conclusions

The results obtained in the present study revealed a possibility to use pine bark as a filler in PLA composites. Composites with large filler particles were characterized by generally better mechanical and physical properties compared to composites with small particles of the filler.

It is noteworthy that both PLA composites with 40\% content of bark had the best parameters. However, the increase in bark content from 40 to 60\% in PLA composites resulted in a decrease of strength.

| Source of variation | TS 2 h | 24 h | WA 2 h | 24 h |
|---------------------|--------|------|--------|------|
|                      | p      | P(\%)| p      | P(\%)|
| a                   | 0.022  | 1.36 | 0.000  | 27.97 |
| b                   | 0.000  | 23.73| 0.000  | 5.39  |
| c                   | 0.000  | 31.39| 0.000  | 27.51 |
| a \times b          | 0.005  | 2.05 | 0.014  | 1.15  |
| a \times c          | 0.000  | 8.16 | 0.003  | 2.28  |
| b \times c          | 0.000  | 4.41 | 0.000  | 5.87  |
| a \times b \times c | 0.020  | 2.07 | 0.000  | 10.17 |
| Error               | –      | 26.85| –      | 19.68 |

\( \alpha \) matrix type/ composite, \( \beta \) size of bark particles, \( \gamma \) share of bark-filler, \( p \) probability of error, \( P \) percentage of contribution

Figure 4 Projection of the variables on the factor plane.
properties of the boards, wherein the decline was most significant in the case of composites with smaller bark particles. The thickness swelling and water absorbability increased as the filler content in the PLA composites increased, and a similar phenomenon was observed for the contact angle and surface energy.

In general, PLA composites were characterized by higher strength parameters and lower humidity resistance in comparison with HDPE composites with the same bark content. Furthermore, PLA composites showed greater surface contact angles with lower total surface energy values.

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Author contributions

PB helped in investigation, conceptualization, methodology, formal analysis, writing—original draft, project administration; funding acquisition. PB helped in investigation, conceptualization, contribution to writing of original draft. RA helped in investigation, conceptualization, contribution to writing of original draft, formal analysis. AA contributed to formal analysis, visualization, writing—review and editing and LD involved in investigation, contribution to writing of original draft. MN and KR investigated the study.

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Data and code availability

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Compliance with ethical standards

Conflict of interest The authors declare that they have no conflict of interest.

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