Advances in Science and Technology Research Journal

Advances in Science and Technology Research Journal, 2026, 20(2), 494–503
https://doi.org/10.12913/22998624/213203
ISSN 2299-8624, License CC-BY 4.0

Received: 2025.10.15
Accepted: 2025.12.06
Published: 2025.12.23

Investigation of the influence of natural fillers on the properties of the samples obtained using additive manufacturing technology

Marta Beata Krawczyk[1*], Rafał Przybylski[2], Paweł Figiel[1], Konrad Kwiatkowski[1]

[1] Faculty of Mechanical Engineering and Mechatronics, West Pomeranian University of Technology, al. Piastow 19, 70-310 Szczecin, Poland
[2] Graduate of the West Pomeranian University of Technology in Szczecin, Poland
* Corresponding author's e-mail: mkrawczyk@zut.edu.pl

ABSTRACT

Natural materials are attracting the interest of researchers as reinforcements in polymer composites because of their low cost, light weight, good mechanical properties, and biodegradability. Numerous studies are being conducted, focusing on the search for the composites that integrate the use of various natural materials as reinforcement with their potential applications in different areas of industry. Polymer composite materials are typically produced using classic methods; however, with the development of manufacturing techniques, they have also become a subject of research in the field of additive manufacturing. This study examined the effect of natural fillers in the form of fibers and powders on thermal degradation, density, water absorption, shrinkage, tensile strength, and impact strength. The samples were made using additive manufacturing with the extrusion technology – MEX (Material Extrusion) at fixed process parameters for each material. The analysis included commercially available materials with a polylactic acid (PLA) matrix at 40 vol.% addition of wood, bamboo, and cork, whereas the results were compared with those of the pure material. The studies showed that the applied natural fillers reduced the density (max. 6%) and tensile strength (approximately 30%), but improved the impact strength (up to 45%). In turn, water absorption increased significantly due to the presence of natural fillers (up to 5.8 times more on the first day). Furthermore, the materials containing natural fillers decompose faster, reducing the thermal stability of the composites.

Keywords: fused filament fabrication, materials extrusion, polymer, natural filler, fiber.

INTRODUCTION

In recent years, there has been growing interest in the biodegradable materials capable of replacing petroleum-based materials, primarily because of their renewable biological origin. With the development and widespread adoption of additive manufacturing methods, it has become essential to utilize these materials in that field as well. One of the most commonly used materials in MEX (Material Extrusion) methods is polylactic acid (PLA), which is utilized in the form of filament [1]. This polymer is characterized by excellent mechanical, thermal, barrier properties, transparency, low cost, and commercial availability [2–4]. As a result, it is widely used across many sectors [5], such as in the packaging industry [6], medicine [7], automotive [1], construction [8–10], as well as in additive manufacturing [11–17]. Despite its advantages, PLA is a brittle material with low impact strength, which may impose certain limitations on its use [18]. Polylactide (PLA) is the most extensively studied biodegradable thermoplastic made from renewable resources for industrial applications [19].

Filling PLA with natural additives (NF) has been widely practiced and researched by scientists at various research centers. For example, Yang et al. [2] studied the feasibility of obtaining films from the PLA doped with cellulose nanocrystals (CNC) by pre-mixing 1 wt% CNC with PLA or with the PLA grafted with glycidyl methacrylate (GMA) (g-PLA). They obtained promising results regarding thermal stability, tensile strength, and modulus of elasticity, all of which were significantly improved compared to those of pure PLA. The conducted research suggests that cellulose-based composite materials with PLA may be used as food packaging materials in various sectors. The PLA-based nanocomposite films with Ag-NPs and thymol were studied in a previous study [20]. The resulting materials demonstrated antioxidant and antibacterial properties with controlled thymol release, thus showing great potential for food preservation and extending shelf life in packaging applications.

Meanwhile, Grassi et al. [21] used the PLA filled with wood fiber for the construction of large-scale structures. Studies have shown that an appropriate content of natural fillers can positively influence durability and the retention of properties during accelerated aging. The type and composition of natural fillers play a key role in determining the characteristics of PLA composites. For example, it has been shown that using natural rubber (NR) as a toughening agent in PLA blends increases ductility, although the degree of improvement largely depends on the fill pattern used during 3D printing [22]. Similarly, the addition of sawdust and soybean oil as post-printing fillers in PLA composites can improve their structural strength and load-bearing capacity [23].

Among other applied and studied natural fillers, the following can be most widely distinguished: flax fibers [15, 24], cocoa bean shell waste [13], wood flour [12, 14, 25], bamboo, and cork [12, 26], also at the nanoscale [6, 20, 27]. The 3D printing materials containing bamboo, cork, and wood have been the subject of research [28, 29]. However, in the first case, the self-made materials contained only powdered natural fillers at a concentration of 30 wt%, and their research also included the materials obtained by injection molding. In the second case, commercial materials with 15 wt% natural phases were used. In both cases, the addition of natural powders had a negative impact on the strength properties of the manufactured materials.

This study focused on the commercially available materials containing a defined 40 wt% amount of natural filler, which, in addition to aesthetic advantages, may also exhibit properties different from those of pure PLA. The main objective of this study was to analyze the effect of natural fillers on various properties, including mechanical properties, density, and water absorption, in relation to pure PLA. The thermal decomposition of individual materials and the effect of natural phases on shrinkage were also investigated. It is worth emphasizing that the integration of natural fillers used in PLA-based materials for 3D printing applications represents a promising step towards more sustainable and environmentally friendly materials. While the improvement in mechanical properties may vary depending on the type and composition of the fillers, the overall benefits in terms of cost reduction and biodegradability are significant [16, 18, 30]. Ongoing research and development in this field are essential to overcome the existing challenges and fully realize the potential of PLA composites with natural fillers.

MATERIALS AND METHODS

Materials

In this study, materials in the form of PLA-based filament from the company print-me (PPHU POLIGRAF Wiesław Kasprowiak, Gorzów Wielkopolski, Poland) were used. Pure polylactic acid filament was used as the reference material. The following filled materials were used: PLA filled with approximately 40 vol.% bamboo particles, PLA filled with approximately 40 vol.% cork fibers and particles, and PLA filled with approximately 40 vol.% wood fiber and powder. To simplify, the sample abbreviations presented in Table 1 were used in this study:

An FFF (Fused Filament Fabrication) device – the Anycubic i3Mega S printer (Anycubic, Shenzhen, Guangdong, China) – as well as the following process parameters, were used: 100% line infill, filament melting temperature 220 °C, bed temperature 60 °C, layer height 0.2 mm, printing speed 50 mm/sec.

A series of samples (n=5) was produced for testing density, water absorption (cubes measuring 20 × 20 × 20 mm), tensile strength, and impact resistance. The variability in the sample was indicated using the standard deviation based on the following equation:

$$\sigma = \sqrt{\frac{\sum_{i=1}^{n}(x_i - \bar{x})^2}{n-1}} \tag{1}$$

where: σ is the population standard deviation; x_i is each individual data point in the population; $\bar{x}$ is the population mean; n is the total number of data points in the population.

Table 1. Samples designation

Sample	Abbreviation
Pure PLA	PLA
PLA + bamboo	BPLA
PLA + cork	CPLA
PLA + wood	WPLA

Physical characterization

Thermogravimetric measurements were carried out on a TG-DSC Q600 device (TA instruments, DE, USA). The samples were heated to 600 °C at a heating rate of 10 K/min in an atmosphere of synthetic air.

Density measurements were performed according to the EN ISO 845:2009 standard, which specifies the method for determining the apparent overall density of porous plastics and rubber. The measurements were performed on 20 samples (five from each series). Before testing, the samples were conditioned by storing them for 72 hours in dry conditions. according to the material data sheets provided by the manufacturer, each material should have a density of 1.2 g/cm^3.

The apparent total density (p) of the samples was calculated using the following formula:

$$p = \frac{m}{V} \times 10^6 \left[\frac{kg}{m^3}\right] \quad (2)$$

where: m is the mass of the sample [g], while V is the volume of the sample [mm^3].

Water absorption was determined by the method of immersion in cold water, following PN-ISO 8361-1:1994. Cubic samples prepared for density measurements were used in the tests, with 5 samples tested for each type of material. Initially, the samples were dried for 24 h at 50 °C, then weighed and placed in a vessel with distilled water at 21 °C for 32 days. Every 24 hours, the samples were removed, dried, and then weighed again. Shrinkage was determined according to the EN ISO 294-4:2019-03 standard as follows: before each measurement, the samples were conditioned under dry conditions for 72 h.

Microscopic examination of the materials fractures after the tensile test was conducted using a 4K Keyence VHX-7000 microscope (Keyence, Osaka, Japan).

Mechanical studies

The impact resistance of the notched specimens was tested using the Charpy method in accordance with the PN-EN ISO 179-1 standard. A Charpy-type apparatus equipped with a 2 J energy hammer was used for this method. Standardized samples (4 × 10 × 80 mm) with a type A notch were used. Tensile tests were performed according to EN ISO 527-1:2020-01 and EN ISO 527-2:2012 standards using an INSTRON ElectroPuls E10000 machine. Type 1A specimens were used in this study. The following parameters were applied: stretching speed of 5 mm/min and jaw distance of 50 mm. Before measurement, the samples were conditioned for 72 h at a temperature of 23 °C and a humidity of 50% (ISO 291:2008).

RESULTS AND DISCUSSION

As part of this research, a series of measurements were conducted, including thermogravimetric analysis (TGA), differential scanning calorimetry (DSC), water absorption testing, tensile strength, impact strength, density, and shrinkage measurements. The results obtained for the composite materials were compared to pure PLA.

TGA-DSC measurements

The analysis was carried out on filaments in the as-delivered condition. Figure 1 shows the mass change curves (TG), derivative of mass change (DTG), and heat flow (HF) obtained from thermogravimetric measurements.

When analyzing the results obtained, it can be observed that the degradation of the tested filaments occurs within the range of 200 to 500 °C and is two-step for those with natural fillers, as indicated by the two stages of mass loss starting at 275 and 405 °C. The resulting curve profiles were characteristic of those recorded for pure natural phases [26, 31]. The addition of NF shifted the PLA decomposition peak towards lower temperatures [8, 13, 15, 31], reflecting the differences in the composition of natural fillers. The degradation of fibers occurs at higher temperatures, which can be observed as an emerging endothermic peak in the temperature range of approximately 420–520 °C. Suberin decomposition occurred in the CPLA sample, whereas lignocellulose decomposition occurred in the BPLA and WPLA samples

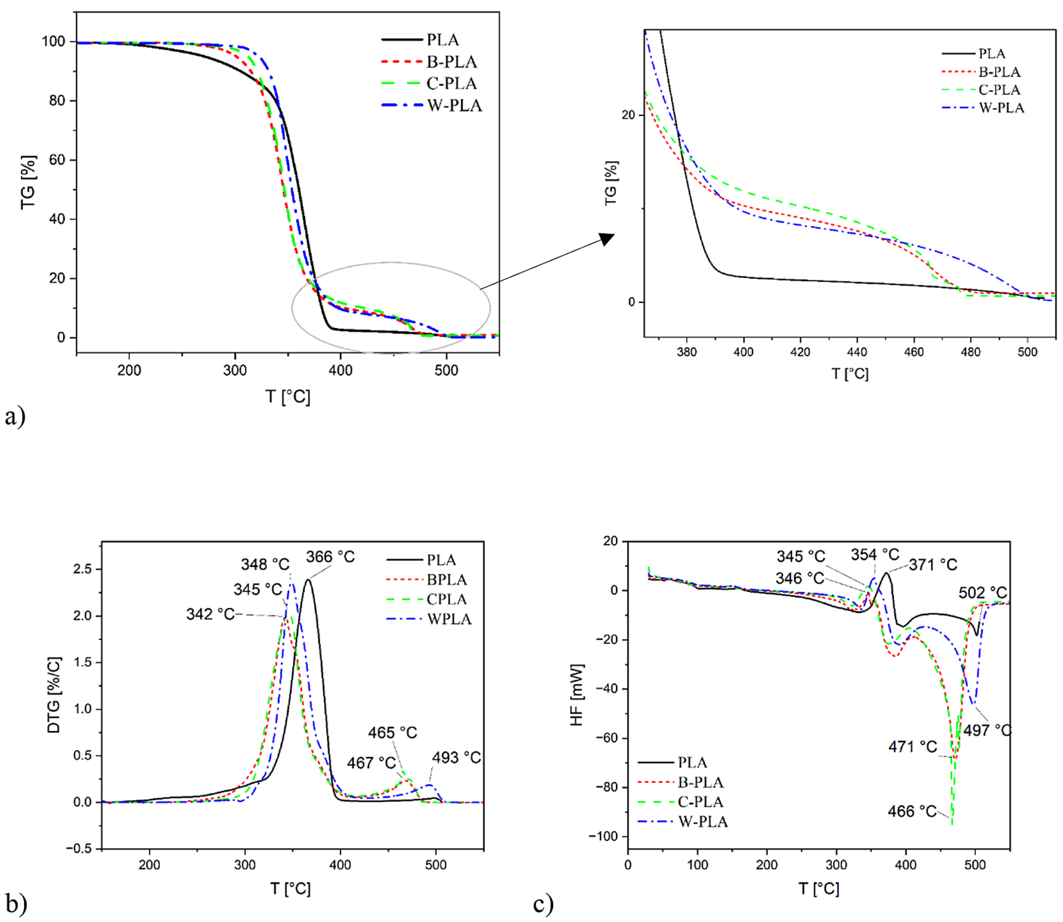

Figure 1. TGA-DSC measurement results: a) mass loss, b) derivative of mass change, c) heat flow

[32]. It is worth noting that the manufacturer does not provide the full chemical composition of the filaments; thus, there may be differences in the additives used, which affect the printability of the filaments and, consequently, the course of the recorded curves. These differences were noticeable at higher temperatures. The provided charts show that the materials with natural fillers degrade more quickly (Figure 1b), which is consistent with the observations of other researchers [8, 13, 15, 18, 31], who also noted that as the proportion of NF increases, the thermal stability of the composites decreases [18]. Bamboo and cork decompose at similar temperatures, whereas wood decomposes at a slightly higher temperature. In all the analyzed samples, similar ranges of enthalpy for phase transitions were observed, consistent with previous studies [15]. In the TG curve, a slope in the temperature range of 375–520 °C can be observed, which does not occur for pure PLA and is attributed to the changes associated with the degradation of natural additives. The mass loss in all samples was approximately 98%, which is consistent with the observations of other researchers [33].

Density measurements

The obtained bulk density measurement results and the theoretical values provided by the material manufacturer are presented in Figure 2.

When analyzing the obtained results, it can be observed that the lowest density was achieved for the samples made from material with the addition of bamboo. With regard to pure PLA, the decreases in the density of the individual materials were as follows: for BPLA, approximately 6.3%; for CPLA, 4.43%; and for WPLA, 4.23%. However, relative to the value specified by the manufacturer, these differences are higher and also apply to the pure material used: PLA – 5.16%; BPLA – 11.15%; CPLA – 9.37%; WPLA – 9.18%. This may be due to the nature of the process, in which the presence of pores and spaces between individual paths of the deposited material is possible, influenced, among other factors, by the parameters used in the 3D printing process. In general, however, it can be stated that owing to natural fillers, it is possible to reduce the mass of the part by as much as around 6%, which may have a significant impact when producing

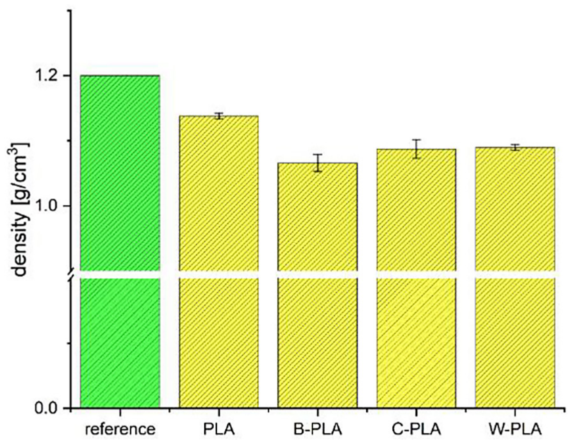

Figure 2. Results of bulk density measurements

large-scale products. This is consistent with the research conducted in other centers [7, 14]. In addition to the NF additives, printing defects – which were also observed by Pereira [34] – could have influenced density. When considering the standard deviation, it is more than three times higher for the materials containing the addition of bamboo and cork. This may be due to the uneven distribution of the natural phase in the material or greater differences in the dimensions of the printed samples.

Water absorption

The behavior of the composites and pure PLA after exposure to water is shown in Figure 3.

When analyzing Figure 3, it can be observed that pure PLA exhibited the lowest water absorption, with a maximum value of 1.5% by weight, due to their hygroscopic properties Materials with the addition of natural fillers showed higher absorbency. This is due to the lignocellulose and suberin content, which are susceptible to water absorption owing to their hydrophilic nature. As early as the first day, the absorption was approximately ~2% for BPLA, ~3.5% for CPLA, and ~4 wt% for WPLA, eventually reaching maximum values ranging from 7.5% for BPLA and CPLA to almost 8.5 wt.% for WPLA after 32 days. Pure PLA absorbed less than 1% by mass of moisture after 24 h, which is consistent with the observations of Banjo et al. [16]. In PLA biodegradation, susceptibility to moisture is the main driving force, which has also been observed in [4, 26, 30, 34, 35], where higher water absorption was also reported for the PLA+NF composites compared to pure PLA, especially in the absence of or with improper matrix modification. However, this may be seen as a potential for the production of elements for sorption drying systems, such as sorption wheels for air drying, as suggested by Martinez-Sanchez et al. in their work [14].

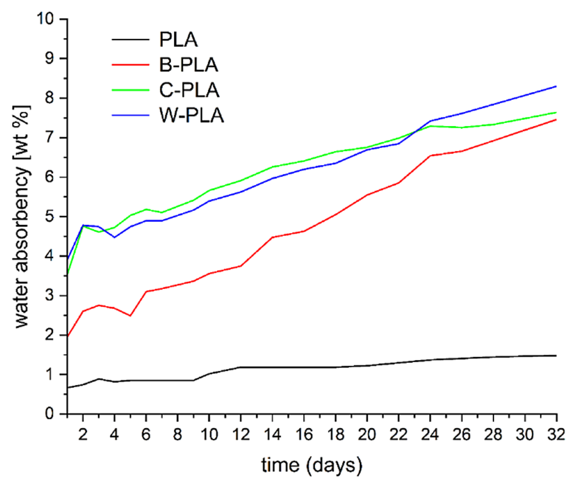

Figure 3. Results of water absorption measurements

Processing shrinkage

According to filament supplier, the volumetric shrinkage should not exceed 0.1%. For the tested materials, this value was significantly exceeded in all cases, as clearly shown in Figure 4.

It can be observed that the highest shrinkage values were recorded for the samples containing bamboo and wood additives. In the case of the cork-containing material, only two samples exhibited shrinkage in the range of 0.85–1.31%, while the remaining samples showed a volumetric increase of up to 0.45%. Considering pure PLA, none of the samples showed shrinkage; rather, an increase in sample volume was noted in the range of 0.7 to 1.35%. The theoretical shrinkage value for PLA should fall between 0.3 and 0.5%. The phenomenon observed may be caused, for example, by excessive extrusion of material through the nozzle, which leads to the expansion of the print paths and consequently increases the dimensions of the final element; due to the relatively small size of the samples, it is possible that, as a result of pressure from the upper layers and the high temperature of the print bed, the first layers may slightly flow outwards, causing an increase in the base dimensions; when the melted filament exits the nozzle, it expands, which may result in a slight increase in the width of the print paths; in the FFF process, the layers cool relatively slowly and not simultaneously, so shrinkage does not

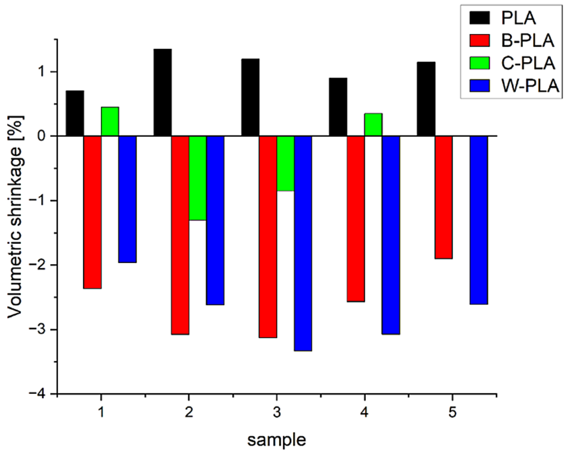

Figure 4. Results of volumetric shrinkage measurements for the tested materials.

occur to the same extent as, for example, during injection molding. Notably, it was observed that both print temperature and layer height affect dimensional accuracy [25]. Therefore, it is possible to reduce the volumetric shrinkage or dimensional increase of the samples through the appropriate selection of these parameters.

Microscopic examination

Figure 5 shows the microscopic images of the fractures obtained for the samples with the addition of natural fillers at 200x magnification.

In the BPLA (Figure 5a) and WPLA (Figure 5c) samples, both fibers and the natural filler powder were visible. The measured fiber diameter for BPLA ranged from 20 to 50 μm, while for WPLA it ranged from 30 to 90 μm. For the CPLA sample, no fibers were observed, which suggests that the main filler phase is the powder, which, if not properly surface-treated chemically, can result in limited interfacial adhesion in the composite and may negatively affect the strength properties of the material, as observed in [36].

Tensile strength tests

The purpose of the test was a comparative analysis of the tensile strength of the samples printed from PLA material with natural fillers such as wood, cork, and bamboo, and a comparison of the results to a part made from pure material. The obtained measurement results are presented in Table 2 and Figure 6.

According to Table 1, the obtained Young's modulus values for each tested material did not reach the values defined by the manufacturer: for pure PLA it was lower by about 26%, for BPLA lower by about 26%, for CPLA by about 30%, and for WPLA lower by about 31%. The material demonstrating the greatest tensile strength is pure PLA, while the lowest tensile strength is observed in PLA with added cork. In general, the addition of natural fillers reduces the tensile strength of PLA-based materials [28, 33, 34, 37, 38], especially at higher filler contents [4]. However, this occurs primarily in the case of unmodified PLA matrices. With the appropriate addition of a compatibilizer or modifier, the presence of natural additives not only does not negatively affect the mechanical properties of PLA-based composites, but actually improves them [33]. Arrigio et al. [39] in their work also emphasized that the final properties of PLA-based biocomposites can be favorably tuned by selecting the appropriate content of natural fillers and the appropriate processing method, as also noted by Mokhena et al. [27]. It is worth mentioning that the results obtained are significantly better than those achieved by Muller et al. [29], who also focused on commercially available materials; this may be due to a higher content of the natural phase or improved bonding between phases. The elongation at break also decreases with the addition of NF to pure PLA, as reported in the literature [8, 11, 28, 34], which leads to increased brittleness of the composites

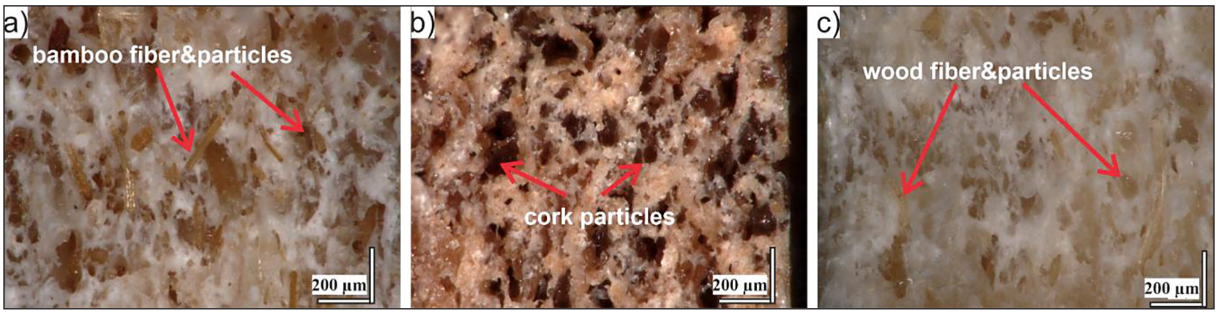

Figure 5. Microscopic images of the fracture of samples: a) BPLA; b) CPLA; c) WPLA at 200x magnification

Table 2. Summary of results for the tensile test. Manufacturer's details are given in brackets

Parameter	Young's modulus/ GPa	Relative elongation at break/ %	Stress at yield point/ MPa	Stress at break/ %
PLA	2.64 (3.50)	6.2	43	29
BPLA	1.95 (3.21)	3.8	24	25
CPLA	1.85 (1.91)	1.8	21	21
WPLA	1.82 (2.72)	3.4	23	24

compared to that of the pure polymer [4]. The main causes for the decrease in Young's modulus can be attributed to the uneven distribution of particles within the composite structure, insufficient extrusion, and defects [34], or inappropriate modification of the PLA matrix [18]. Mechanical properties can also be improved through chemical treatment of the fibers, which has a positive effect on load transfer at the bio-filler-polymer interface [4, 10, 36]. It has been observed that pure PLA exhibits better mechanical properties than composite samples, while the selection of an appropriate compatibilizer improves interfacial adhesion and positively influences the results of strength tests [13]. Figure 6 presents an exemplary tensile diagram obtained for a BPLA sample (a) and a comparative diagram for the tested materials (b).

The courses shown in Figure 6b indicate that, apart from PLA, the other tested materials exhibit brittle behavior – after reaching the maximum force value, they undergo a sudden failure without a plastic phase, known as "necking", especially in the case of C-PLA.

The results obtained are consistent with those reported by other researchers. It has been observed that in order to achieve higher strength properties, it is better to use continuous natural fibers [4, 24]. Pure PLA has a continuous, homogeneous structure that transmits stress well and provides better adhesion between layers during 3D printing. The natural fibers added to PLA do not form chemical bonds with PLA, which weakens the structure of the material and introduces discontinuities that may become crack initiation points. Therefore, the prints made from such materials are more brittle and less resistant to tensile stress. A higher tensile strength of a material indicates a greater ability to withstand tensile loads without damage or breaking, which is crucial in the applications exposed to such stresses. Therefore, when strength properties are most important, a pure PLA filament without additives will work best.

Impact test

According to the impact test results presented in Figure 7, all of the composites exhibited higher impact strength values compared to pure PLA

The observed trend is identical to that obtained by the manufacturer. The material with the highest energy absorption capacity turned out to be PLA with cork admixture (an increase of about 45%), while the lowest was pure PLA. For the remaining materials, the recorded increase ranged between 6% and 15%. Therefore, it can be stated that the addition of cork, bamboo, and wood decreased the tensile strength but improved the impact resistance, which is consistent with the observations of

a) b)

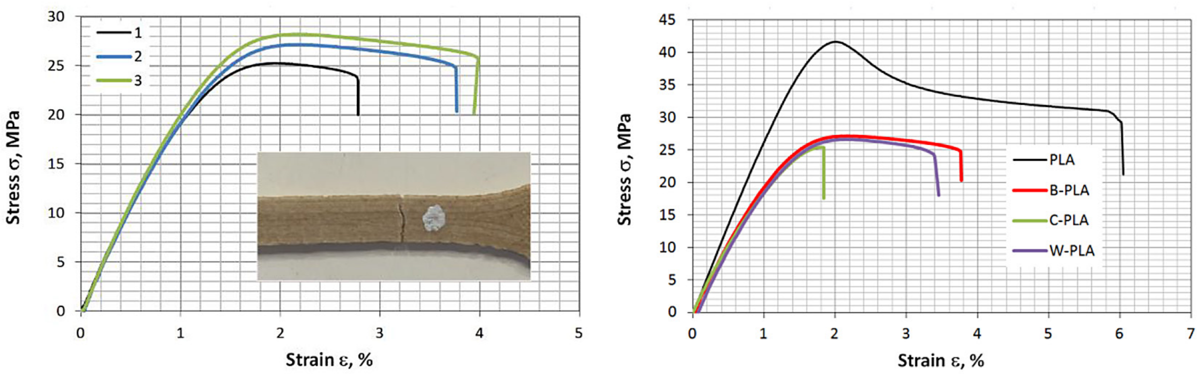

Figure 6. Example stress- strain diagram obtained for BPLA samples (a) and for the tested materials (b)

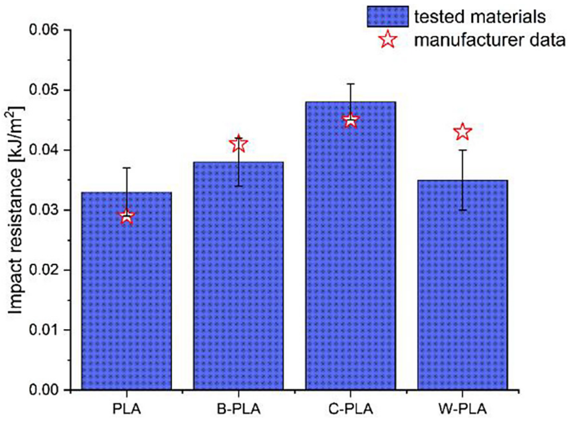

Figure 7. Results of Charpy impact tests. Manufacturer's data are marked with an asterisk

other researchers [23, 31]. The addition of natural particles/fibers caused the material to no longer be homogeneous, but instead become a composite. In such material, the particles acted as micro-springs or points that prevented crack propagation. During impact, energy disperses at the PLA-fiber phase boundaries, resulting in micro-displacements instead of a single, sharp fracture. However, the obtained results differ from those reported for experimental materials [28] and described in [4], where it was found that the impact strength of PLA always decreased after adding natural fibers or particles. This may be due to better homogenization of the natural phase in the material or better selection of additive manufacturing process parameters. The results of the conducted studies indicate that natural additives affect not only the appearance of the object produced by additive techniques, but also its strength parameters. Taking standard deviation into account, it can be observed that the results are consistent and similarly concentrated around the average value for all materials. It is worth noting that higher impact resistance of plastics indicates a greater ability to absorb impact energy, which can be desirable in the applications where protection against mechanical damage is important, such as casings, covers, or wall panels. Therefore, when these properties are essential, a PLA filament with a cork additive is the best choice.

CONCLUSIONS

The aim of the study was to examine the influence of natural fillers on the properties of PLA-based composite materials used in the additive manufacturing process. Thermal behavior, density, shrinkage rate, moisture adsorption, tensile strength, and impact strength were investigated. The obtained results were compared to those of pure material and the manufacturer's data. The main conclusions from the study are as follows:

- The addition of natural fillers (NF) in the form of bamboo, cork, and wood in the amount of 40% vol. reduces the density of the resulting materials by over 6%.
- The tested biocomposites demonstrated reduced thermal stability compared to the pure PLA matrix.
- NFs have a positive effect on the dimensional behavior of the samples. In the case of BPLA and WPLA, greater dimensional stability was observed than for the CPLA material and pure PLA.
- The materials with added NFs exhibited up to five times higher affinity for water adsorption compared to the pure matrix.

Tensile strength decreases in the presence of NFs; however, higher strength was achieved for the materials also containing chopped fibers.

The materials with a natural phase addition are able to absorb more energy in the Charpy impact test, with CPLA exhibiting the highest value (45% more than that recorded for PLA).

By comparing the obtained results with the manufacturer's catalogue data, it can be concluded that none of the tested materials achieved the defined strength parameter values. This may be caused, for example, by the use of different manufacturing process parameters. However, it is worth emphasizing that natural additives have a positive effect on density (reducing it by up to 6%) and impact strength (increasing it by up to 45% in the case of cork) compared to pure PLA. Unfortunately, they also have a negative effect on tensile strength (deteriorating it – reducing Young's modulus by up to 31% in the case of wood), water absorption (up to 8 wt.%), and shrinkage (increasing it to as much as 3%).

Therefore, further research should focus on selecting the manufacturing process parameters for each material to improve the achieved mechanical properties. Studies on resistance to degradation under varying environmental conditions are also planned.

It can be stated that the tested biodegradable PLA-based materials are a promising trend in the 3D printing market. Not only do they visually resemble natural base materials, but owing to their different properties, they open up new possibilities for application.

Acknowledgements

Microscopic examinations were conducted using a 4K Keyence VHX-7000 device purchased as part of project number: RPZP.01.03.00-32-0001/21 Upgrading of the Environmental Measurement Laboratory of the Faculty of Mechanical Engineering and Mechatronics in the laboratory of intelligent materials and damping structures. The project was co-financed by the European Union from the European Regional Development Fund under the Regional Operational Programme for the West Pomeranian Province 2014–2020.

REFERENCES

1. Ciofu C., Mazurchevici S.-N., Maldonado-Cortes D., Pena L., Correa D. I. Q., Nedelcu D. Tribological behavior of PLA biodegradable materials used in the automotive industry. Materials 2020; 13(16): 3615. https://doi.org/10.3390/ma13163615
2. Yang W., Dominici F., Fortunati E., Kenny J. M., Puglia D. Melt free radical grafting of glycidyl methacrylate (GMA) onto fully biodegradable poly(lactic) acid films: effect of cellulose nanocrystals and a masterbatch process. RSC Adv. 2015; 5(41): 32350–32357. https://doi.org/10.1039/C5RA00894H
3. Garcia-Campo M. J., Quiles-Carrillo L., Sanchez-Nacher L., Balart R., Montanes N. High toughness poly(lactic acid) (PLA) formulations obtained by ternary blends with poly(3-hydroxybutyrate) (PHB) and flexible polyesters from succinic acid. Polym. Bull. 2019; 76(4): 1839–1859. https://doi.org/10.1007/s00289-018-2475-y
4. Mazzanti V., Malagutti L., Mollica F. FDM 3D Printing of polymers containing natural fillers: A review of their mechanical properties. Polymers 2019; 11(7): 1094. https://doi.org/10.3390/polym11071094
5. Tümer E. H., Erbil H. Y. Extrusion-based 3D printing applications of PLA composites: A review. Coatings 2021; 11(4): 390. https://doi.org/10.3390/coatings11040390
6. Ainali N.M., Tarani E., Zamboulis A., Pusnik Cesnar K., Fras Zemljic L., Chrissafis K., Lambropoulou D.A., Bikiaris D.N. Thermal stability and decomposition mechanism of PLA nanocomposites with Kraft Lignin and Tannin. Polymers 2021; 13(16): 2818. https://doi.org/10.3390/polym13162818
7. Fouly A., Assaifan A., Alnaser I., Hussein O., Abdo H. Evaluating the mechanical and tribological properties of 3D printed polylactic-acid (PLA) green-composite for artificial implant: Hip Joint Case Study. Polymers 2022; 14(23): 5299. https://doi.org/10.3390/polym14235299
8. Bhagia S., Bornani K., Agrawal R., Satlewal A., Ďurkovič J., Lagaňa R., Bhagia M., Yoo Ch.G., Zhao X., Kunc V., Pu Y., Ozcan S., Ragauskas A. J. Critical review of FDM 3D printing of PLA biocomposites filled with biomass resources, characterization, biodegradability, upcycling and opportunities for biorefineries. Applied Materials Today 2021; 24: 101078. https://doi.org/10.1016/j.apmt.2021.101078
9. Grassi G., Lupica Spagnolo S., Paoletti I. Fabrication and durability testing of a 3D printed façade for desert climates. Addit Manuf 2019; 28: 439–444. https://doi.org/10.1016/j.addma.2019.05.023
10. Ilyas R.A., Sapuan S.M., Harussani M.M., Hakimi M.Y.A.Y., Haziq M.Z.M., Atikah M.S.N., Asyraf M.R.M., Ishak M.R., Razman M.R., Nurazzi N.M., Norrrahim M.N.F., Abral H., Asrofi M. Polylactic acid (PLA) biocomposite: Processing, additive manufacturing and advanced applications. Polymers 2021; 13(8): 1326. https://doi.org/10.3390/polym13081326
11. Fico D., Rizzo D., De Carolis V., Esposito Corcione C. Bio-composite filaments based on poly(lactic acid) and cocoa bean shell waste for fused filament fabrication (FFF): Production, characterization and 3D printing. Materials 2024; 17(6): 1260. https://doi.org/10.3390/ma17061260
12. Bulanda K., Oleksy M., Oliwa R., Budzik G., Gontarz M. Biodegradable polymer composites based on polylactide used in selected 3D technologies. Polimery 2020; 65(07/08): 557–562. https://doi.org/10.14314/polimery.2020.7.8
13. Brounstein Z., Yeager C. M., Labouriau A. Development of antimicrobial PLA composites for fused filament fabrication. Polymers 2021; 13(4): 580. https://doi.org/10.3390/polym13040580
14. Martínez-Sánchez J. A., Romero P. E., Comino F., Molero E., Ruiz De Adana M. Effect of material extrusion process parameters to enhance water vapour adsorption capacity of PLA/wood composite printed parts. Polymers 2024; 16(20): 2934. https://doi.org/10.3390/polym16202934
15. Belarbi Y. E., Benmahiddine F., Hamami A. E. A., Guessasma S., Belhabib S. Hygrothermal and Microstructural Investigation of PLA and PLA-Flax Printed Structures. Fibers 2022; 10(3): 24. https://doi.org/10.3390/fib10030024
16. Banjo A. D., Agrawal V., Auad M. L., Celestine A.-D. N. Moisture-induced changes in the mechanical behavior of 3D printed polymers. Composites Part C: Open Access 2022; 7: 100243. https://doi.org/10.1016/j.jcomc.2022.100243
17. Sharma S., Majumdar A., Butola B.S. Tailoring the biodegradability of polylactic acid (PLA) based films and ramie- PLA green composites by using selective additives. Int J Biol Macromol 2021; 181: 1092–1103., https://doi.org/10.1016/j.ijbiomac.2021.04.108

18. Yu M., Zheng Y., Tian J. Study on the biodegradability of modified starch/polylactic acid (PLA) composite materials. RSC Adv. 2020; 10(44): 26298–26307. https://doi.org/10.1039/D0RA00274G
19. Kfoury G., Hassouna F., Raquez J., Toniazzo V., Ruch D., Dubois P. Tunable and durable toughening of polylactide materials via reactive extrusion. Macromol Mater Eng 2014; 299(5): 583–595. https://doi.org/10.1002/mame.201300265
20. Ramos M., Beltran A., Fortunati E., Peltzer M., Cristofaro F., Visai L., Valente A.J.M., Jiménez A., J.M. Kenny, Garrigós M.C. Controlled release of thymol from poly(Lactic Acid)-Based Silver Nanocomposite Films with Antibacterial and Antioxidant Activity. Antioxidants 2020; 9(5): 395. https://doi.org/10.3390/antiox9050395
21. Grassi G., Lupica Spagnolo S., Paoletti I. Fabrication and durability testing of a 3D printed façade for desert climates. Additive Manufacturing 2019; 28: 439–444. https://doi.org/10.1016/j.addma.2019.05.023
22. Fekete I., Ronkay F., Lendvai L. Highly toughened blends of poly(lactic acid) (PLA) and natural rubber (NR) for FDM-based 3D printing applications: The effect of composition and infill pattern. Polymer Testing 2021; 99: 07205, https://doi.org/10.1016/j.polymertesting.2021.107205
23. Ramisetty Y. T., Schuster J., Shaik Y. P. Investigating the impact of 3D printing parameters on hexagonal structured PLA+ samples and analyzing the incorporation of sawdust and soybean oil as post-print fillers. JMMP 2024; 8(5): 193. https://doi.org/10.3390/jmmp8050193
24. Le Duigou A., Barbé A., Guillou E., Castro M. 3D printing of continuous flax fibre reinforced biocomposites for structural applications. Materials & Design 2019; 180: 107884. https://doi.org/10.1016/j.matdes.2019.107884
25. Chaidas D., Kechagias J. K. An investigation of PLA/W parts quality fabricated by FFF. Mater. Manuf. Processes 2022; 37(5): 582–590. https://doi.org/10.1080/10426914.2021.1944193
26. Mofokeng J. P., Luyt A. S., Tábi T., Kovács J. Comparison of injection moulded, natural fibre-reinforced composites with PP and PLA as matrices. J. Thermoplast. Compos. Mater. 2012; 25(8): 927–948. https://doi.org/10.1177/0892705711423291
27. Mokhena T. C., Sefadi J. S., Sadiku E. R., John M. J., Mochane M. J., Mtibe A. Thermoplastic processing of PLA/cellulose nanomaterials composites. Polymers, 2018; 10(12): 1363. https://doi.org/10.3390/polym10121363
28. Bulanda K., Oleksy M., Oliwa R., Budzik G., Gontarz M. Biodegradable polymer composites based on polylactide used in selected 3D technologies. Polimery 2020; 65(07/08): 557–562. https://doi.org/10.14314/polimery.2020.7.8
29. Müller M., Šleger V., Kolář V., Hromasová M., Piš D., Mishra R. K. Low-cycle fatigue behavior of 3D-printed PLA reinforced with natural filler. Polymers 2022; 14(7): 1301. https://doi.org/10.3390/polym14071301
30. Scaffaro R., Maio A., Gulino E. F., Alaimo G., Morreale M. Green composites based on PLA and agricultural or marine waste prepared by FDM. Polymers 2021; 13(9): 1361. https://doi.org/10.3390/polym13091361
31. Daver F., Lee K. P. M., Brandt M., Shanks R. Cork–PLA composite filaments for fused deposition modeling. Compos. Sci. Technol. 2018; 168: 230–237. https://doi.org/10.1016/j.compscitech.2018.10.008
32. Nirmal Kumar K., Dinesh Babu P. Experimental study of mechanical, morphological and thermal performance of bamboo-reinforced polylactic acid-based montmorillonite clay hybrid composite. Ind Crops Prod 2024; 209: 117950. https://doi.org/10.1016/j.indcrop.2023.117950
33. Vinod A., Tengsuthiwat J., Vijay R., Sanjay M. R., Siengchin S. Advancing additive manufacturing: 3D-printing of hybrid natural fiber sandwich (Nona/Soy-PLA) composites through filament extrusion and its effect on thermomechanical properties. Polym. Compos. 2024; 45(9): 7767–7789. https://doi.org/10.1002/pc.28302
34. Pereira D. F., Branco A. C., Cláudio R., Marques A. C., Figueiredo-Pina C. G. Development of composites of PLA filled with different amounts of rice husk fibers for fused deposition modeling. J. Nat. Fibers 2023; 20(1): 2162183. https://doi.org/10.1080/15440478.2022.2162183
35. Nawadon P., Wasinee C., Phisut N., Borwon N. 3D printing filaments prepared from modified poly(Lactic Acid)/teak wood flour composites: An investigation on the particle size effects and silane coupling agent compatibilisation. Phys. Sci. 2019; 30(2): 169–188. https://doi.org/10.21315/jps2019.30.2.10
36. Dobrosielska M. et al. Influence of diatomaceous earth particle size on mechanical properties of PLA/diatomaceous earth composites. Materials 2022; 15(10): 3607. https://doi.org/10.3390/ma15103607
37. Sabbatini B., Cambriani A., Cespi M., Palmieri G. F., Perinelli D. R., Bonacucina G. An overview of natural polymers as reinforcing agents for 3D printing. ChemEngineering 2021; 5(4): 78. https://doi.org/10.3390/chemengineering5040078
38. Piš D., Pouzarová H., Hanušová K. Degradation of 3D printed polymer composites with filler of cellulose-based materials. Manuf. Technol. 2022; 22(3): 327–333. https://doi.org/10.21062/mft.2022.041
39. Arrigo R., Bartoli M., Malucelli G. Poly(lactic Acid)–Biochar Biocomposites: Effect of processing and filler content on rheological, thermal, and mechanical properties. Polymers 2020; 12(4): 892. https://doi.org/10.3390/polym12040892