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The PLA content influence selected properties of wood-based composites

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Abstract: *The PLA content influence selected properties of wood-based composites.* The aim of the research was to find how the lower (25%) and higher (50%) PLA content affects the selected properties of the obtained WPC samples. The result of the strength tests (compressive strength) shows, that the increasing content of PLA, increases the compressive strength value. The ash content value determining mineral saturation, decreasing with increasing PLA content but there is no significant difference. The moisture content was carried for wood sample by drying the material and was figured by differences in the material weight mass. Raw material samples was compacted inside the chamber to form the composites by using high temperature (230°C) and strength not higher than 600 N. Optical analyzed was performed for analyzing the structure of the prepared sample, and comparing that structure before and after strength test. It can be concluded, that increasing thermoplastic content in biocomposites causes better strength, and it has not significant bad impact on the environment.

Keywords: Wood-Plastic Composites, polymer, poly (lactid acid), plastic, wood chips

INTRODUCTION

Wood-Plastic Composites, WPC for short (Chmielnicki et al. 2017), are a strongly growing branch of wood-based plastics production. These composites include wood flour and synthetic polymers, together forming the base (Wnorowska et al. 2017). There are various methods of manufacturing WPC products, the choice of which depends on the subsequent use of the prepared material and the assumed properties in terms of strength and aesthetics. These properties depend not only on the manufacturing method but also on the type of wood and polymer used. The degree and method of grinding and the influence of admixture of other components also have an impact. Polymer-wood composites are characterized by high resistance to fungi and bacteria due to their low absorbency and less sensitivity to external agents than wood (Zajchowski et al. 2005). The fillers of WPC composites are waste materials from the wood industry. Most often they are in the form of wood flour, dust, chips, fibers or sawdust. Wood flour is formed by grinding wood in several stages. It has lipo- and hydrophilic properties, is insoluble in water, and its composition depends on the kind of tree. The warp of the composites are polymers. Thermoplastics in WPC make the processing easier. They improve resistance to moisture and to decomposition of wood (Chmielnicki et al. 2017). Warp materials should have a melting temperature of no more than 200°C (Postawa et al. 2003). Differentiation of composite materials (except their subsequent use) is also made by their construction. The properties of the recived composites, are therefore depend on the content of wood chips. There is a classification into three groups. The first is low-filled composites, with a small wood chips content, 10-40% by weight. The second contains 40-80% by weight of wood chips, and these are high-filled composites. And in the third, where the wood chips mass content reaches 90%, are composites called "liquefied wood" (Zajchowski and Ryszkowska 2009).

WPC processing techniques are extrusion and injection molding methods that do not require the need for additional machining. Otherwise, typical tools such as woodworking machines are enough. During production processes, composites can be colored in the mass, or surface of finished products can be finished by painting, laminating or veneering (Zajchowski and Tomaszewska 2008). The manufacturing process also includes compression molding and hot pressing. For the composite, the important factors are melting temperature and pressure, which selection must not cause thermal decomposition of the wood (Gardner et al. 2015). Processing usually includes three steps. The first is filler processing, which is milling, screening and drying. The next stage is mixing the filler (for example, wood flour) with the polymer warp. The final step is the molding of finished products, using the previously mentioned methods (Włodarczyk-Fligier et al. 2018).

Processing properties are specifically determined by the measurement of stickiness changes. Stickines are a measure of the resistance to the flow of plasticized materials. In the study presented below, it was measured using a capillary rheometer for WPC mills of various compositions, which had previously been plasticized in a crushing machine adding wood flour, and then cooled and grinded. It was observed that with the increase of filler content, the plasticized composite stickiness increasing in comparison to the polymer material which is the warp (Lewandowski et al. 2014).

The softwood specifications are different for juvenile wood (lower) and mature wood (higher). This dependency was confirmed by studies, which involved measuring the compressive strength along the fibers, bending strength , the value of the strength quality factor in compression and bending of the wood. In the course of the analyses, the distance from the trunk base was considered and showed that the properties studied, along with the conventional density, decrease in relation to growth, where the more dynamic character is shown by mature wood. As a result, it was observed that the biggest differences in parameter values between juvenile and mature wood are closer to the trunk (Tomczak and Jelonek 2012).

The wood of the pine (*Pinus sylvestris* L.) (Krzysik 1975, Tomczak and Jelonek 2012), common to the softwood type, is resinous. It has light yellow and wide sapwood and red-brown heartwood. The latewood zone is clearly visible, wide. Annual rings are also clearly visible. The most important technical features include easy to process, splittability, durability, the wood is medium-heavy and has medium mechanical properties (Krzysik 1975). The feature that negatively affects the technical value is inhomogeneity, which is at the same time an advantage from a biological point of view, while the way the raw material is used is of major importance in this aspect (Tomczak et al. 2009).

Thermoplastics are polymers from the plastomer group (technological classification). This group is characterized by small deformations (up to 1%) under tension and the fact that when they are exposed to increasing loads, they show plastic deformations until mechanical damage is caused. Thermoplastics are plastics that soften when heated, they can then be shaped. They harden and regain their original properties when cooled, maintaining the shapes they were previously given (tworzywa.pwr.wroc.pl). They consist of linear or branched macromolecular chains (macromolecules consist of large molecules formed after joining together to form chains of small molecules) that are not connected to each other. These plastics can be thermoformed, which allows for the removal of cracks and features (Bajerlejn et al. 2017). Thermoplastics can be formed many times without changing their physical-mechanical properties (Waśniewski 2016).

One of the biodegradable aliphatic polyesters is poly(lactic acid) PLA for short (Farah et al. 2016). This biodegradable polymer is made from recyclable materials (Georgiopoulos et al. 2015). PLA is not toxic or harmful and does not cause problems when it comes to the environment. This material is produced from plants such as corn and potato creating a more environmentally friendly thermoplastic. The material is characterized by high elasticity and plasticity, which increases as the raw materials melt flow temperature is raised to 190 - 230°C. Commercially, this composite is easily available in stores specializing in the sale of components and plastics for 3D printers. Its advantage is the environmentally friendly process of material formation (Borysiewicz 2018). The study, in which WPC samples composed of thermally modified or acetylated wood components (wood flour) and PLA, examined the biological durability, sorption and mechanical properties of the injection molded samples. Tests showed that the composites and clean PLA were very slow to absorb moisture. When biological durability was tested, no significant results were found, while strength losses were noted after moisture sorption treatment, from which it can be concluded that compatibility between wood particles and PLA should be improved (Feng et al. 2020).

The aim of the study was to identify the influence of the additive content of PLA (25% and 50%) in the production of WPC from wood chips, on the properties of the obtained composites (compressive strength, ash content, moisture content) and their statistical analysis.

MATERIALS AND METHODS

Materials

The study required pre-treatment of wood raw material and PLA. The material used in the study was shredded with a fraction not exceeding 1 mm. The tests gave been conducted according to PN-EN 15149-1:2011 and PN-EN 15149-2:2011 standards. The test takes 120 seconds, where the fractions of 0 ÷ 1 mm were separated from the test material. The same shredding procedure was applied for the wood and PLA test material. After fragmentation material was mixed according to the weight. Thanks to the preparation of materials in accordance with the purpose of the work, a mix of 25% and 50% PLA with wood was prepared as a base for further research.

Samples preparation

In order to carry out the planned tests, the compaction head was used. An appropriate test measuring stand was constructed to prepare the specimens for strength testing. The prepared raw material sample was compacted inside the chamber at the desired temperature. According to the recommendations during preparation, the controller maintained a constant temperature of the heaters. Required temperature in this case obtain in 230°C. Compaction included a one-time compression with a strength not higher than 600 N. It was assumed that if the structure of the produced sample does not obtain a satisfying quality, other force values will be taken. The compaction process was made for samples with a volume about 9.5 cm3. The constant parameter was the inner diameter (Ø11 mm), and the length of the compaction chamber (100 mm). The parameters of the compaction head, as well as the maximum compaction strength (600 N) determined the maximum unit pressure, whose value reaches 19.83 MPa.

Strength tests

The prepared samples were studied according to the strength tests. For studies of strength tests the Instron machine 3382, with an attachment suitable for testing was used. The testing machine was plugged to computer as a part of the measuring stand. The study concerned the determination of compressive strength of prepared specimens (Roman et al. 2021a). The purpose of this test is to measure the characteristics from which the failure stress, yield strength and strain energy of a specimen can be determined (Roman et al. 2019). The static compression test was conducted according to the standards of PN-D-04102:1979; PN-D-04229:197727; PN-D-04115:195828.

Optical analyze

The optical analyze involved a scanning method characteristic using the Nikon SMZ 1500 microscope and the computer program for data analizes. This stereoscopic microscope has zoom range 15:1 and the range of zooms from 0.75 to 112.5. This allows to see samples from macro views to high-magnification micro visualization. The microscope is equipped with an optical system with full correction of chromatic aberration - planar and axial. Its halogen illuminator has an output of 150 W.

Picture desaturation

Computer image editing allows converting a source image into a final image form. Image transformation involves permanently changing colors, by converting RGB type to grayscale. Changing an image's color to grayscale causes irreversible data loss, meaning that the RGB mode cannot be recovered.

For image analysis, the GIMP (GNU Image Manipulation Program) v. 2.20.32 program was used. The algorithm in a specially prepared program counted the shades of gray to analyze the pixel colors. The computer program interface was designed and programmed in the Delphi environment.

Moisture content

The moisture content of the shredded prepared mix samples was tested according to the guidelines of PN-ISO 589:2006. The moisture content of the prepared material was additionally controlled before compaction. The tests were carried out with the use of the Radwag WS30 type weighing-dryer.

Ash content

The ash content determines mineral saturation, as its composition includes silicon, iron, aluminum, calcium, magnesium, sodium, potassium (Babiarz, Bednarczuk 2013). The measurement of ash content was performed according to the method of slow ashing in a muffle oven (Kotlicki 2007). The weighed sample was placed in a crucible of known mass. The weights used for the study, prepared in triplicate (with an instrument measurement accuracy of 0.0001 g), had a mass of about 3 g per sample. The samples were ashed in a muffle oven, which had to be warmed to 805°C before testing.

Statistical Analysis

Statistical analysis of the study results was performed using ANOVA analysis of variance (Roman et al. 2021b). The method allowed us to determine the influence of parameters of shredded wood material (pine) and PLA thermoplastic on the mechanical strength of the resulting product. During the statistical analysis, other factors were assumed to be invariant (i.e. ceteris paribus).

RESULTS AND DISCUSSION

The composite's pro-ecological also manifests itself in the ash content. During the process, organic macronutrients such as carbon (C), hydrogen (H), oxygen (O), nitrogen (N), and sulfur (S) are burned, determining the ash content. The ash content determines the mineral saturation of the tested raw material, as silicon, iron, aluminum, calcium, magnesium, sodium, and potassium can be specified in its content (Babiarz, Bednarczuk 2013). The percent content of mineral compounds and moisture content of the measured raw material for the native samples and mixtures used in all the tests are summarized in Table 1.

Table 1. Mineral compounds and moisture content**.**

Proportion of PLA - pine mix (%)	Moisture (%)	Ash content (%)
100 – 0	-	1.362
50 – 50	11.31	1.377
25 – 75	12.06	1.414
0 – 100	11,96	1.426

After ashing, the material took on a different color depending on the ashed material. The ash from PLA was intensely white. With 50% wood content intensely white ash had a shade of gray. The ash content of 25% PLA was dominated by a gray color with additive white ash. The wood ashed into typical gray ash. It was observed that as PLA decreased, the ash content in percent increased. This trend is presented in Figure 1.

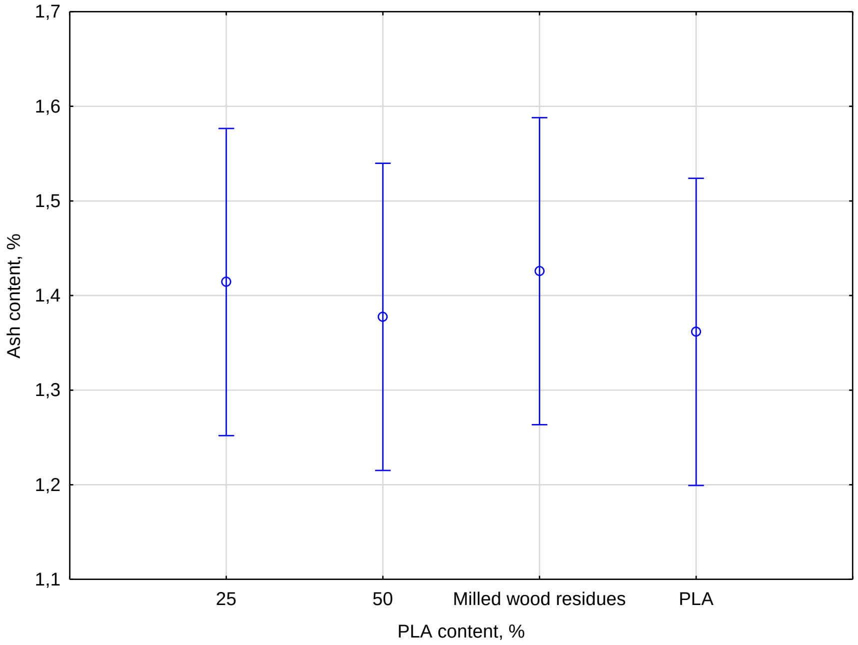

Figure 1. The PLA content influence ash content

Despite the fact that an increase in wood content in the biocomposite resulted in an increase in ash content of an average value of 0.064%, statistical analysis and the Dunkan post-hock test performed didn't show a significant difference. The obtained results were statistically processed by conducting an analysis of variance to determine the degree of significance of the tested parameter. During the statistical analysis, it was found that the degree of significance had a value of $p = 0.90412$ and was higher than the accepted critical level. The empirical value of the statistic was $F(3, 8) = 0.18430$.

The prepared samples were studded under the microscope to analyze the structure of the side and above view, with 25 and 50% of PLA. In the first place was noted, that the structure of the samples was different according to the PLA share. Both cases were presented in figures 2 and 3.

(**a**) (**b**)

Figure 2. The sample of 25% PLA share: (**a**) Side view; (**b**) View from above

(**a**) (**b**)

Figure 3. The sample of 50% PLA share: (**a**) Side view; (**b**) View from above

The image characteristics were for overview to provide information on the composition of the different fractions. In the photo of the sample with less PLA, less compactness in the structure is seen. Wood chips in the sample where they have a higher content (in figure 2) make the surface darker and rougher. PLA is less visible, in the form of a thin layer of light-colored mass.

In figure 3, the PLA is more visible, in the form of a thicker mass, with a color in the white tone. Gives the impression of a cohesive material, making the wood chips more structured, compacted into a mass. The surface texture is smoother.

After microscope analysis, the samples were studied in Instron tested machine. To make the assumption that allows comparing the samples in the later time, the surface displacement during the static compression was 5mm/min of speed. The graph interpretation of the average static compression test for the prepared samples was presented in Figure 4.

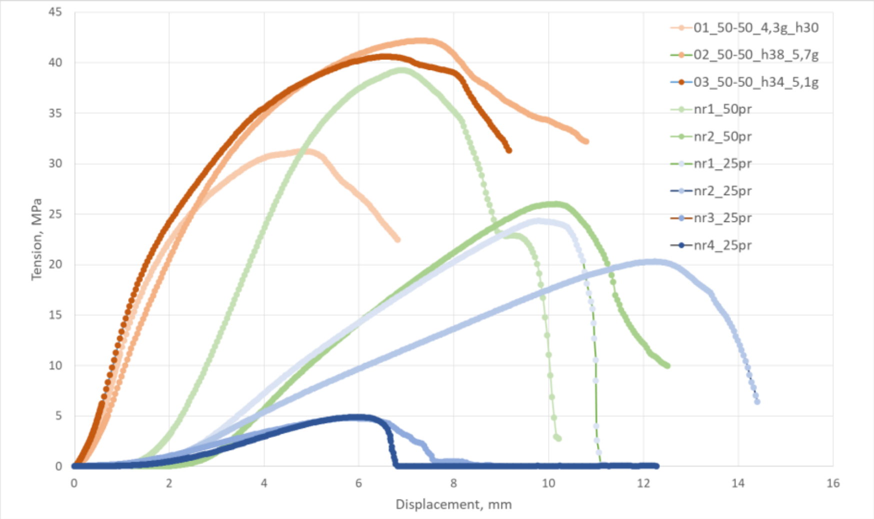

Figure 4. The average static compression test for the prepared samples

Measured changes in compaction strength at different raw material and process parameters, were performed according to the methods on an Instron testing machine and integrated computer program. The average value of biocomposites parameters was presented in table 2.

Table 2. The average value of biocomposites

Proportion of PLA - pine mix (%)	Average tension (MPa)	Average density (kg/m^3)
50 – 50	35.85	953.06
25 – 75	13.58	735.61

The ANOVA analysis allows determining the influence of individual factors on the output parameter, by homogeneous group classification, and the mutual relationships of the analyzed parameters as well. To qualify a data set to one homogeneous group, the critical level of significance determining assignment should be below 0.05 (5%). The expected boundary averages are presented in Figure 5.

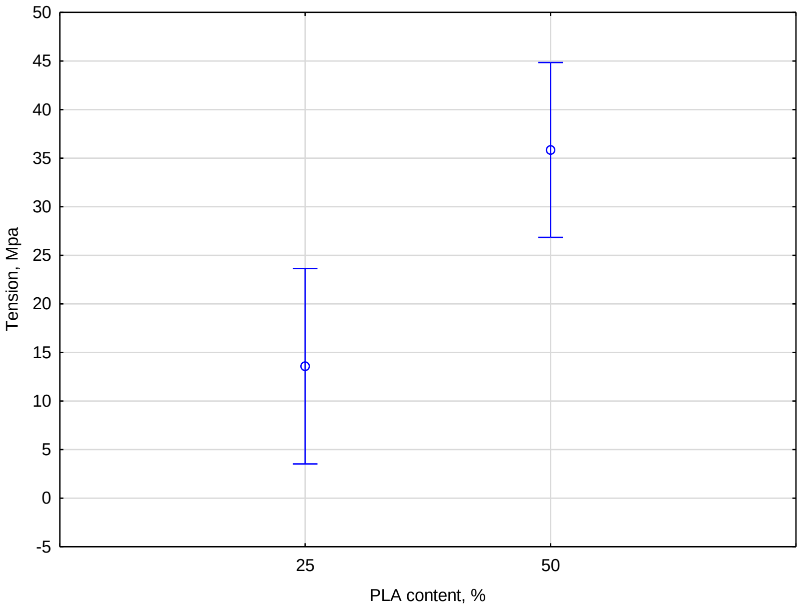

Figure 5. Tension value in relation to the raw material mixture

Statistical analyze allowed to characterize the results in view the influence of the raw material mixture on the strength of the tested biocomposite. The performed statistical analyze showed the differences between the measured parameters. The results determine the value of the degree of significance of $p = 0.00586$ for the empirical value of the statistic $F(1, 7) = 15.246$. According to the method, the post-hoc analysis using Duncan's test showed a difference between the parameters. The significance degree p was lower than the accepted alpha significance level of 0.05, separating the parameters into two homogeneous groups. A comparison showing the average effects of the parameter influence analyze is presented in Table 3.

Table 3. The influence of the raw material mixture on test biocomposite strength

PLA content (%)	Mean tension (MPa)	Homogenous group	
25	13.58	X	
50	35.85		X

The conducted statistical analyze of the influence of raw material mixture on strength of tested biocomposite showed the existence of two homogeneous groups. This means significance of the influence measured strength parameter of the prepared composite. The biocomposite with 50% PLA had higher strength values by more than half. The average strength of this biocomposite was 35.85 MPa, 22.27 MPa higher than the biocomposite whose mixture contained 25% PLA. This also means that in order to increase the plasticity of the raw material, the PLA content in the mixture should be increased. This observation can be concluded from the crack characteristics of the biocomposite during the compression test. An example of biocomposite cracks depending on the PLA content is shown in Figure 6.

(**a**) (**b**)

Figure 6. The internal structure of the sample: (**a**) 25% PLA share; (**b**) 50% PLA share

The samples after strength tests once again were put under a microscopy analyzes to study the internal structure of 25 and 50% PLA share. It helps to observe influence of the mechanical damage on the composite structure. This photos also allow to compare how the composite structure changes depends on the PLA content.

The material fractions, despite the compressive forces, did not show much deformation. The differences in the structure presented in the microscope photos, between before and after the strength test are not very noticeable. It can be seen that after these tests, the fraction elements are more disorganized, but the wood chips and PLA mostly stay in the same form as before. This was probably a result of grinding the raw material during composite preparation.

The results of the microscopic analyses were statistically analyzed to verify the performance of the program in image characterization of PLA percentage. The analysis assumption was to identify the PLA percentage, based on the desaturated image. Using one-way ANOVA variance analyze compared the program's percentage of PLA per factor characterizing the density of gray tones in the analyzed image read from the computer program. The diagram shows the variation of cases and therefore, the possibility of existing significant differences between the measured parameters. The results from analyzing the influence of PLA percentage on the factor characterizing the density of grayscale color are presented in Figure 7.

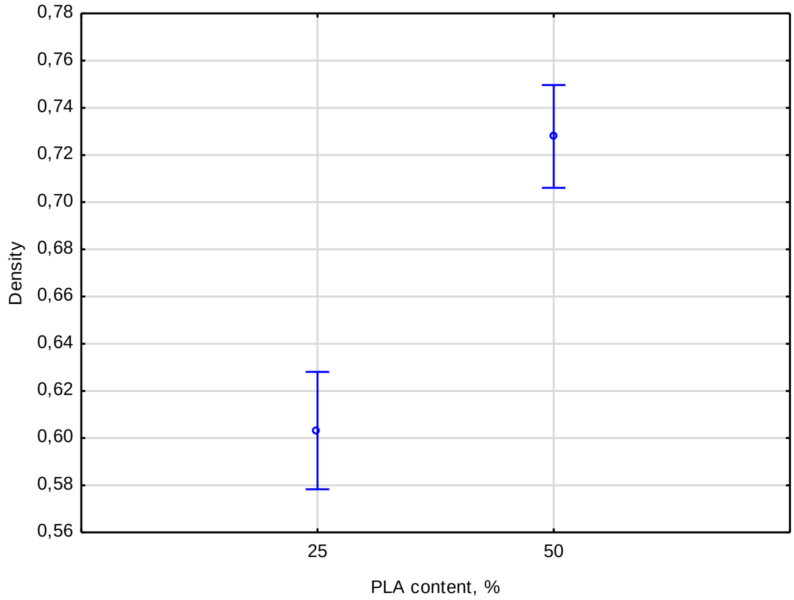

Figure 7. The analysis effects of PLA content on the parameter describing the density of grayscale tones

The established level of significance in the measurement of the influence of the percentage of PLA on the coefficient characterizing the density of gray shades was lower than 0.05, therefore, in accordance with the principle, in order to determine homogeneous groups, the Duncan statistical test was performed. Homogeneous groups defining the affiliation of individual mixtures with the participation of PLA in relation to the density factor are listed in table 4.

Table 4. The influence of the raw material mixture on test biocomposite strength

PLA content (%)	Mean density coefficient	Homogenous group	
25	0.603	X	
50	0.728		X

Statistical analyze allowed to characterize the results in the context of PLA content percentage impact on the factor characterizing color density from grayscale tones. Post-hoc statistical analyzes showed the presence of significant differences between the measured parameters. The results were determined by a significance value of p was less than 0.05, and the empirical statistic F(1, 81) = 56.240.

CONCLUSIONS

The results show, that the increasing content of PLA thermoplastic in WPC has the highest impact on mechanical properties (compressive strength). Ash content values of prepared materials decreasing with increasing PLA content, but does not show the significant difference. Microscope analyze show that PLA helps to bond the composite structure. The PLA additive to wood chips in the WPC manufacturing has a positive effect on the strength properties, plasticity and appearance of the material. It can be concluded that it is a good compound for composites, considering its biodegradability, because this will not be a danger to the environment.

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Streszczenie: *Wpływ udziału PLA na wybrane właściwości kompozytów drewnopochodnych*. Celem badań było ustalenie, w jaki sposób niższa (25%) i wyższa (50%) zawartość PLA wpływa na wybrane właściwości otrzymanych próbek WPC. Wynik badań wytrzymałościowych wskazuje, że zwiększająca się zawartość PLA zwiększa wartość wytrzymałości na ściskanie. Zawartość popiołu określająca nasycenie minerałami zmniejsza się wraz ze wzrostem zawartości PLA, ale nie ma istotnej różnicy. Zawartość wilgoci obliczono dla materiału drzewnego przez suszenie materiału i obliczono na podstawie różnic w masie materiału. Próbki surowców zagęszczano wewnątrz komory w celu uformowania kompozytów przy użyciu wysokiej temperatury (230°C) i wytrzymałości nie większej niż 600 N. Przeprowadzono analizę optyczną w celu analizy struktury przygotowanej próbki i porównania tej struktury przed i po testach wytrzymałościowych. Można stwierdzić, że zwiększenie zawartości tworzyw termoplastycznych w biokompozytach powoduje poprawę wytrzymałości i nie ma istotnego negatywnego wpływu na środowisko.

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