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Flax pomace as a substitute for wood raw material in lignocellulosic composite technology

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Abstract: *Flax pomace as a substitute for wood raw material in lignocellulosic composite technology.* In the present study, composites made from flax seed pomace and PE, PP, PLA and modified starch were subjected quality parameter tests. Density, modulus of rapture (MOR), and modulus of elasticity (MOE) were examined, as well as water absorption and swelling to thickness after 2h and 24 h soaking in water, wettability and surface roughness. Addition of flax to different polymers caused significant changes of parameters but the level of influence of the addition varied between polymers. Although the characteristics of the produced biocomposites have been shown to be in general inferior to pure polymers, efforts should be made to improve these characteristics. Agri-food waste products can be a valuable raw material for the production of new type of biomaterials.

Keywords: biocomposites, plant waste, polyethylene, polylactide, polypropylene, starch, sustainable materials, alternative resources

INTRODUCTION

In 2000, Poland harvested 27.659 mln^3 of timber (Łączyński et al. 2011), while in 2021 this value has increased to 42.24 mln m^3 (CSO 2022). As the population grows and the areas of wood application continue to expand, the demand for wood raw material will continue to rise. To sustain the growth in the wood-based composites industry, the furniture industry, and the construction industry, it is necessary to consider the possibility of adopting lignocellulosic raw materials from other industries, where they constitute challenging-to-utilize production residues.

An example of a product in which wood particles are successfully substituted by other raw materials is wood-plastic composites - WPC. These are materials that combine the advantages of both components. Thanks to the use of synthetic polymers, these composites are resistant to weathering, allowing them to be widely used outdoors under exposed conditions (Yáñez-Pacios and Martín-Martínez 2017). Another significant advantage of WPC composites is that they can be processed using equipment commonly used in wood processing (Zbieć et al. 2010). Wood filler in WPC composites improves the strength properties of these materials and, simultaneously, offers the possibility to reduce their density. In addition, a great advantage of WPC is the possibility to use post-consumer wood and recycled plastic in their manufacturing process (Elsheikh et al. 2022). The main methods of WPC production are extrusion and injection, in which the filler is added in ground form to form fibers (Chmielnicki and Jurczyk 2013). Another developing method of WPC production is extrusion of polymer blends in an extruder into granular form and subsequent secondary flat-pressing using traditional shelf presses. The results of testing the properties of WPC with a density of 750 kg/m^3 and a thickness of 16 mm, produced with 50 % polyethylene (PE) or polypropylene (PP) and 50% wood particles, are known. Plates produced as a result of the extrusion of polymer blends, which were given their final form by flat pressing, were characterized by the following strength parameters: from PE a modulus of rapture of 12.9 MPa, a modulus of elasticity of 1264 MPa, while from PP a modulus of rapture of 20.5 MPa, a modulus of elasticity of 2144 MPa. The role of filler in WPC composites can also be played by lignocellulosic material extracted from plants such as straw, phloem, leaves, seeds and grasses (Borysiuk 2012). As a potential valuable substitute for wood, it is also possible to use agri-food processing by-products such as flax pomace. Oilcakes (a.k.a. pomace) are the residues of seeds after oil pressing. Oilcakes after flax pressing are mainly used as an ingredient in animal feed (Jasinska-Kuligowska 2018). The use of flax pomace as a filler has been studied so far, but these studies involved composites made by extrusion. PP specimens with flax as 30 % by weight were characterized by a tensile strength of 27.3 MPa, a tensile modulus of 1659 MPa, and a modulus of rapture of 42.9 MPa and a modulus of elasticity of 3900 MPa (Arbelaiz et al. 2005). Significantly better results were recorded for composites with 40 % flax and 2 % maleated polypropylene (MAPP), which improved the bonding of the filler particles to the polymer. The static bending strength in this case was 67 MPa, the tensile strength was 42 MPa, and the tensile modulus was 3200 MPa. Only in the case of the static bending modulus value was there a reduction relative to the PP plates (3400 MPa and 3900 MPa, respectively) (Li and Sain 2003).

With reference to the cited results of studies on WPC-type composites, this paper presents the results of studies on selected physical and mechanical properties of lignocellulosic-polymer composites made with flax pomace and four polymers, i.e.: polyethylene (PE), polypropylene (PP), polylactide (PLA) and modified starch (MS).

MATERIALS AND METHODS

Composites were produced using the following raw materials:

1. lignocellulosic particles in the form of flax pomace (AL-PHADAR, Nienaszów, Poland)
2. thermoplastics:
    a. high-density polyethylene - HDPE (PE) (Orlen Polyolefins Sp. z o.o., Płock, Poland),
    b. polylactic acid / polylactyde (PLA) (Resinex Poland Sp. z o.o., Kraków, Poland),
    c. polypropylene (PP) (post-consumer),
    d. modified starch (MS) (Grupa Azoty S.A., Tarnów, Poland).

The first step in the production process was the preparation of raw materials. The lignocellulosic particles were crushed with a hammer mill, after which they were dried to a moisture content of 5 %. The next process was homogenization, i.e. creating a homogeneous mixture of dried particles with polymers, using a high-speed mixer (KMOD SGGW, Warsaw, Poland). After this operation, composed material was produced by extrusion (this procedure was commissioned to be carried out at the Research and Development Center for Wood-Based Panels Ltd. in Czarna Woda), which was then ground in a hammer mill. Homogenization and extrusion of the material were intended to prevent the particles from screening each other and to ensure that the lignocellulosic particles and the polymer in question were evenly distributed throughout their volume. The mixtures ratio was 1:1.

The next step was to produce composites in the form of slabs with an assumed density of 900 $kg/m^{3.}$ The boards were manufactured in the assumed dimensions (length x width x thickness) 275 mm x 160 mm x 2.5 mm. In the case of boards with modified starch as a matrix, water was added in the amount of 9 % of the weight of the mats (a value determined on the basis of technological tests and organoleptic evaluation). The mats were then formed using the circulation sheets of the press and the mold prepared from HDF board. The material was separated from the press circulating sheets with aluminum foil and paper to use eliminate the phenomenon of sticking of mats to the press circulating sheets. The material prepared in this way was subjected to pressing in a single-shelf laboratory press (AB AK Eriksson, Mariannelund, Sweden) at 200 °C, except for samples with modified starch, for which the pressing temperature was set at 180 °C (values determined on the basis of technological trials and organoleptic evaluation). The adopted pressing parameters of the mats followed the scheme shown in Figure 1. The material was then subjected to cooling, also under pressure (press - Industrial Equipment Plant, Nysa, Poland).

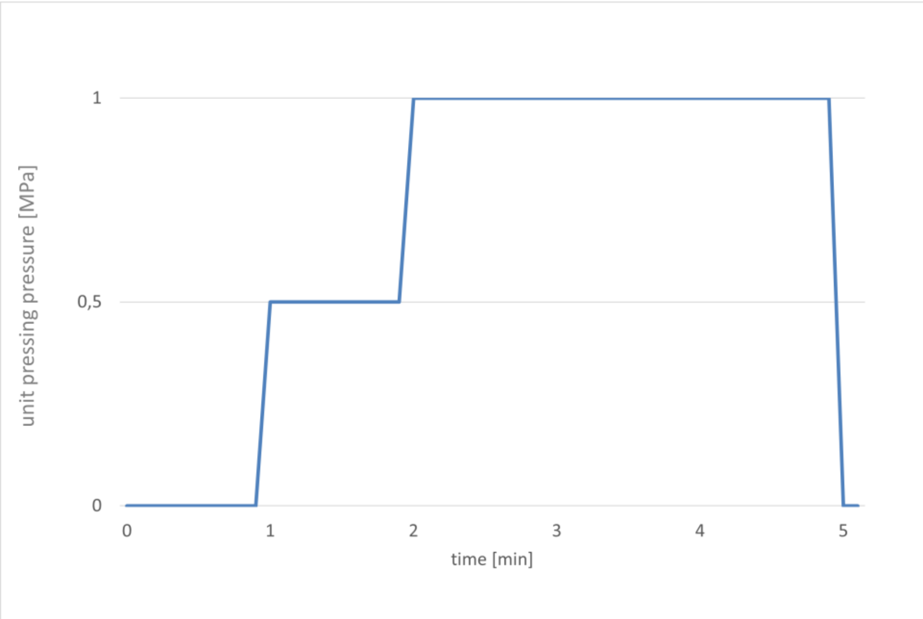

Figure 1. Pressing parameters of the mats

The manufactured boards were subjected to tests to determine their selected properties, i.e.:

- Density according to PN-EN 323:1999 - Wood-based panels - Determination of density.
- Static bending strength and static bending modulus in accordance with PN-EN 310:1994 - Wood-based panels - Determination of bending modulus and bending strength.
- Thickness swelling (TS) and water absorption (WA) after 2h and 24h immersion in water according to PN-EN 317:1999 - Particleboard and fiberboard - Determination of swelling to thickness after soaking in water.
- Water absorption after 2h and 24h soaking in water [%] was calculated according to the formula: $WA_n = (m_n-m_0) / m_0 \cdot 100$ % where: m_0 - mass of sample, m_n – mass of sample after soaking in water, n- time, $n \in (2, 24)$.
- Contact angle using Haas Phoenix 300 goniometer (Surface Electro Optics, Suwon City, South Korea) with controlled, automatic dispensing of 1 µL droplets. To analyze the data, received by the camera, the software - Image XP version 5.8 (Surface Electro Optics, Suwon City, South Korea) - was used, which automatically determines the angle between the edge of the reference liquid (distilled water) droplet deposited on the material surface. The wetting angle was determined 5, 20, 40 and 60 seconds after the reference liquid droplet was deposited on the surface of the test material.
- Surface roughness using Surftest SJ-210 portable contact profilometer (Mitutoyo Co., Kawasaki, Japan), based on the assumptions of PN-EN ISO 21920-2:2022-06 - Product Geometry Specifications (GPS) - Geometric Structure of Surfaces: Profile - Part 2:

Terms, definitions and parameters of geometric structure of surfaces (Rz parameter was defined for each type of material).

At least 10 repetition for each test were performed for every variant of the board. The experimental data was statistically analyzed using STATISTICA 13.3 software. Statistical inference was performed for the significance level α = 0.05. The mean values of determined parameters were examined in the one-way analysis of variance (ANOVA) - Tukey's post hoc test, in which homogeneous groups of mean values for each parameter were identified for p = 0,05.

RESULTS

This section present the results of conducted tests of mechanical and physical parameters. Results for all types of produced composites were compared with parameters of pure termoplastics as a reference samples.

Density:

For all types of thermoplastics, except PLA, the density of the composite was found to be higher than that of the samples without flax. The largest difference of 13 % was shown by samples with PP. The opposite relationship describes the samples with PLA. In this case, it was the density of the thermoplastic that turned out to be 18 % higher than that of the composite.

Table 1. Density of composite variants and control samples (pure polymers: MS, PE, PP, PLA)

Variant	density [kg/m^3]	standard deviation [kg/m^3]	coefficient of variation [%]	a, b, c, d – homogeneous groups by Tukey test (α = 0.05)
FLAX_PLA	874	67.19	5.79%	a
PLA	1061	23.88	2.06%	c
FLAX_PE	957	20.22	1.74%	b
PE	897	4.41	0.38%	a
FLAX_PP	988	20.01	1.72%	b
PP	873	18.33	1.58%	a
FLAX_MS	1161	35.61	3.07%	d
MS	1062	2.23	0.19%	c

Table 2. Thickness of composite variants and control samples (pure polymers: MS, PE, PP, PLA)

Variant	thickness [mm]	standard deviation [mm]	coefficient of variation [%]	a, b, c – homogeneous groups by Tukey test (α = 0.05)
FLAX_PLA	2.53	0.01	0.34%	b
PLA	2.22	0.07	3.23%	a
FLAX_PE	2.40	0.10	4.59%	b
PE	2.59	0.03	1.63%	c
FLAX_PP	2.39	0.06	2.81%	b
PP	2.64	0.10	4.81%	c
FLAX_MS	2.11	0.08	3.95%	a
MS	2.13	0.02	1.12%	a

Mechanical properties

The tests showed a decrease in the strength of materials with flax, relative to plates made of thermoplastics alone. For samples with polyethylene, the difference is small – 11 %

what confirmed Tukey's test. For samples with PLA and PP, which showed much higher strength results, the strength decrease is much greater, at 95 % and 56 %. Such a high, relative to the others, difference in strength of samples from PLA is further influenced by their inverse relationship of density of composites and thermoplastics. Relative to the other composites, which showed a static bending strength of less than 10 MPa, flax with polypropylene stands out. The result of this material is as high as 17.92 MPa.

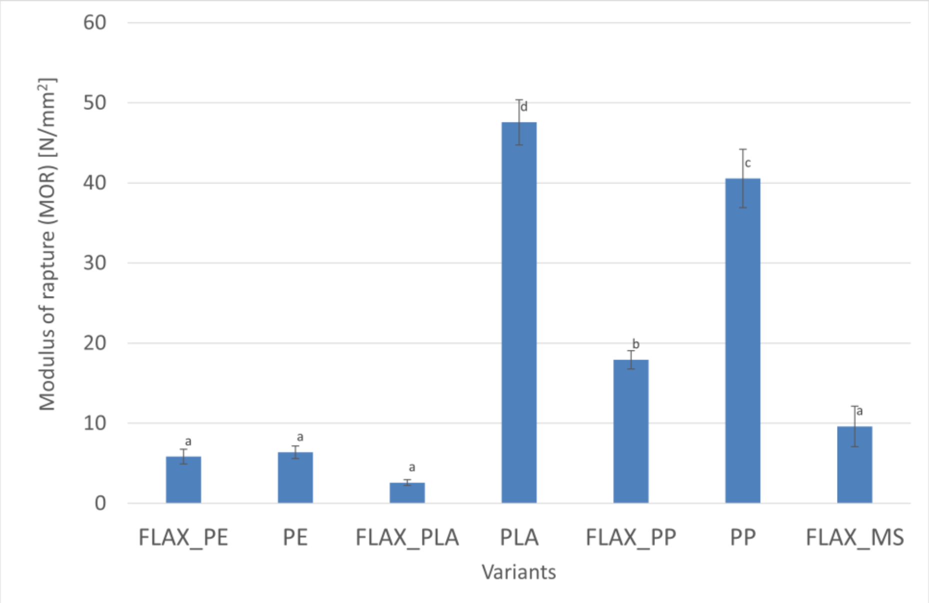

Figure 2. Modulus of rapture of composite variants and control samples (pure polymers: MS, PE, PP, PLA); a, b, c, d – homogeneous groups by Tukey test (α = 0.05)

For all materials, the results were characterized by a large scatter, so extreme results were omitted from the analysis. The highest elastic modulus results were obtained by FLAX_ PP and this samples are in one homogenous group with PP. Other composites with their MOE result are other one homogenous group.

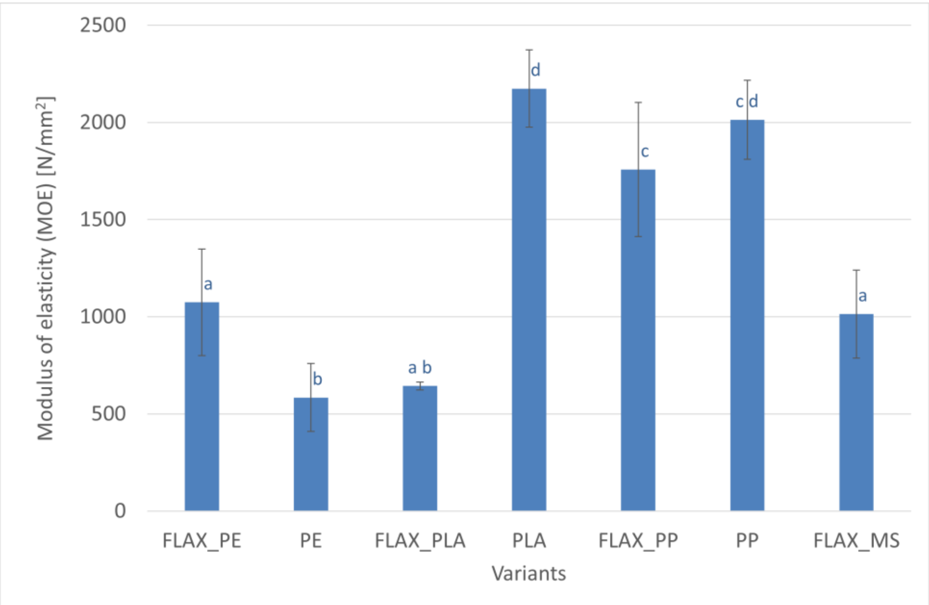

Figure 3. Average values of modulus of elasticity;
a, b, c, d – homogeneous groups by Tukey test (α = 0.05)

Samples made of starch alone were omitted from the study, as these showed significantly different characteristics from the other samples. They were to a significant degree more pliable and elastic. It was not possible to test them on the machine on which the other samples were tested, while the results from another device would not be meaningful.

Water absorption and thickness swelling

Pure PE, PLA and PP has a very low water absorption. Water uptake can be caused by microcracks or in case of post-consumer polymers the presence of some water-absorbent contaminants (Guo et al. 2019). Addition of flax particles cause an increase of water absorption. FLAX_PLA composite stands out from other with almost 60 times higher water absorption after 24h comparing to pure PLA. Materials made of modified starch during the test disintegrated as early as 2 h after the test began.

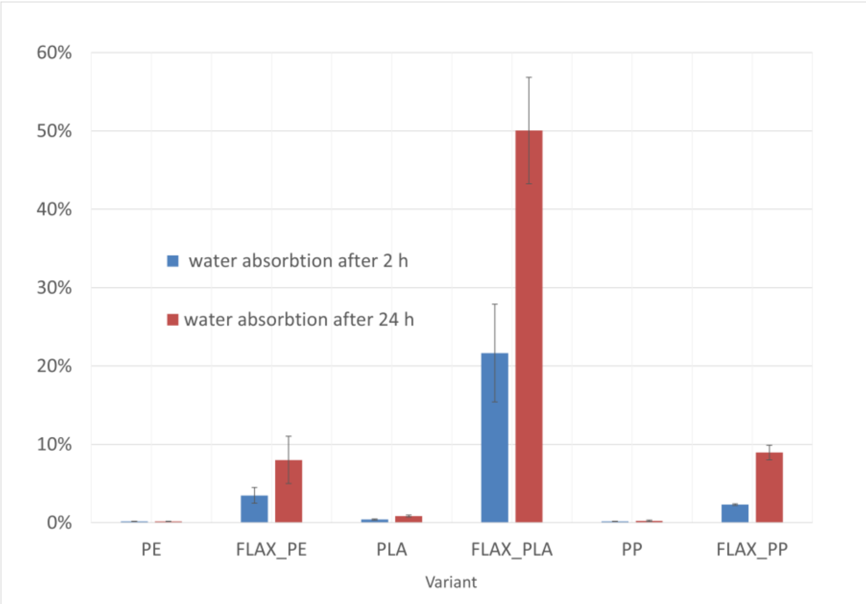

Figure 4. Water absorption after 2h and 24h of composite variants and control samples (pure polymers: MS, PE, PP, PLA)

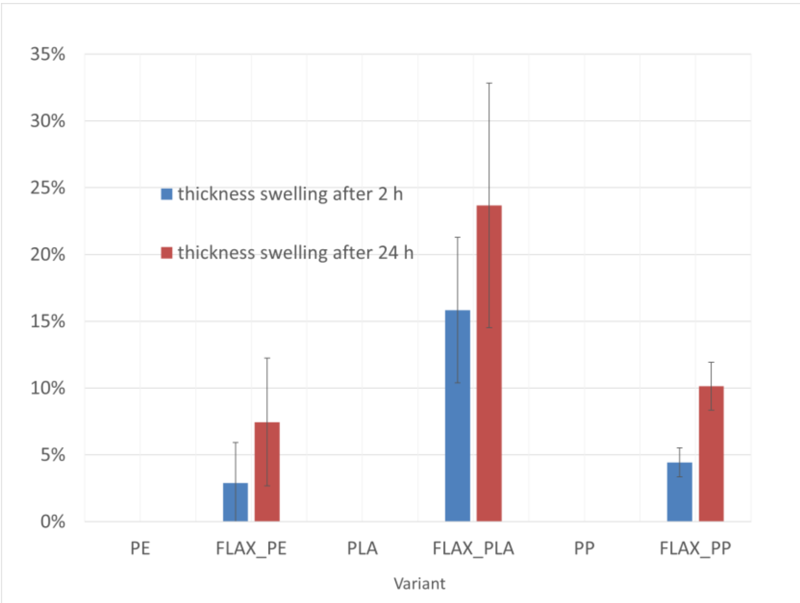

Figure 5. Thickness swelling after 2h and 24h of composite variants and control samples (pure polymers: MS, PE, PP, PLA)

Contact Angle

Wettability test results for FLAX_PP and PP samples were similar. The wetting angle 5 sec. after droplet application was, in the case of the composite, larger, by only 2.7°. All other materials showed a smaller and slower decreasing wetting angle in the form of the composite with flax than as samples made of thermoplastic alone. The largest difference in wetting angle measured after 5 sec between the two was for the PE products - 26.3°. On the FLAX_MS composite, the droplet was the least stable - the wetting angle decreased in 55 sec by as much as 30.3° - after 60 seconds it was only 7.7°, moreover, all the results obtained by this sample were the lowest. Materials that achieve a wetting angle of less than 90° are considered hydrophilic. [Paslawska Ż. 2017]

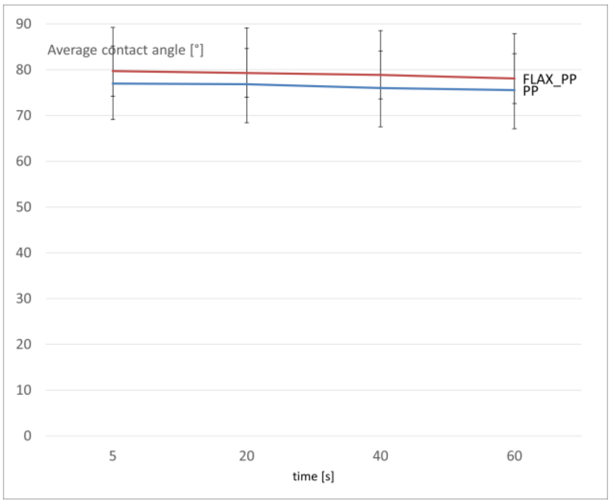

Figure 6. Contact angle values measured 5, 20, 40 and 60 seconds after droplet application of composite variants and control samples (pure polymers: MS, PE, PP, PLA)

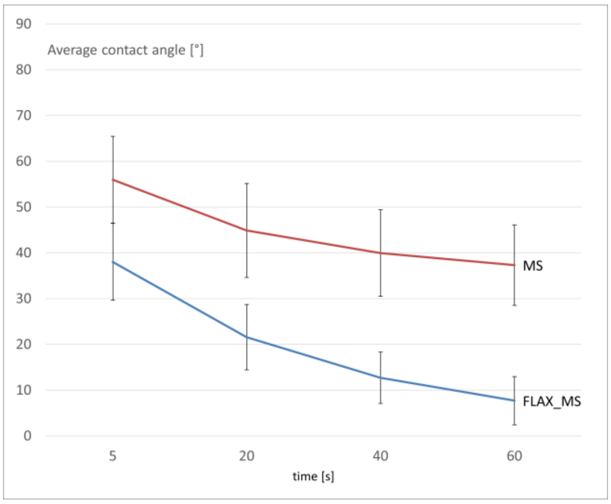

Figure 7. Contact angle values measured 5, 20, 40 and 60 seconds after droplet application of composite variants and control samples (pure polymers: MS, PE, PP, PLA)

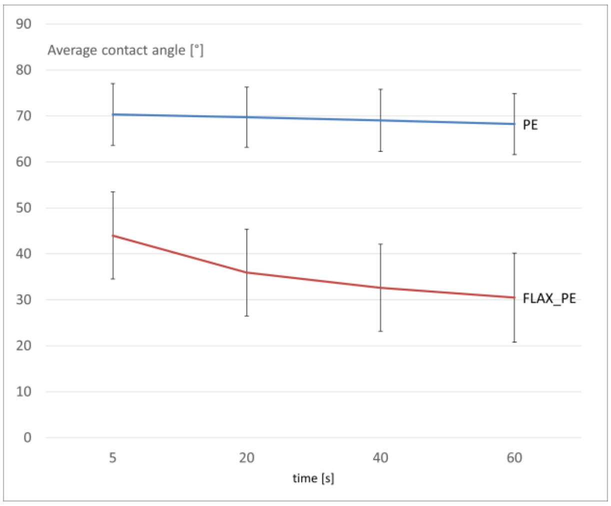

Figure 8. Contact angle values measured 5, 20, 40 and 60 seconds after droplet application of composite variants and control samples (pure polymers: MS, PE, PP, PLA)

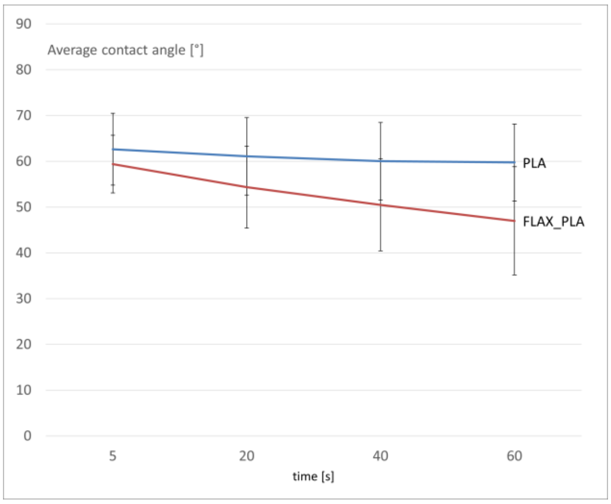

Figure 9. Contact angle values measured 5, 20, 40 and 60 seconds after droplet application of composite variants and control samples (pure polymers: MS, PE, PP, PLA)

Surface roughness

All materials as thermoplastic flax composites obtained higher results of Rz and Ra coefficients than samples made of thermoplastics alone, but according to homogenous groups by Tukey's test, for materials with PLA and PP the difference was not significant. Addition of flax to polymers influenced notably parameters in case of MS and PE. Ra parameter did not exceed in any case 10 μm. If the Ra parameter is between 1.25 and 10 μm, the material is considered moderately rough. [Maritime University of Gdynia - Faculty of Mechanical Engineering].

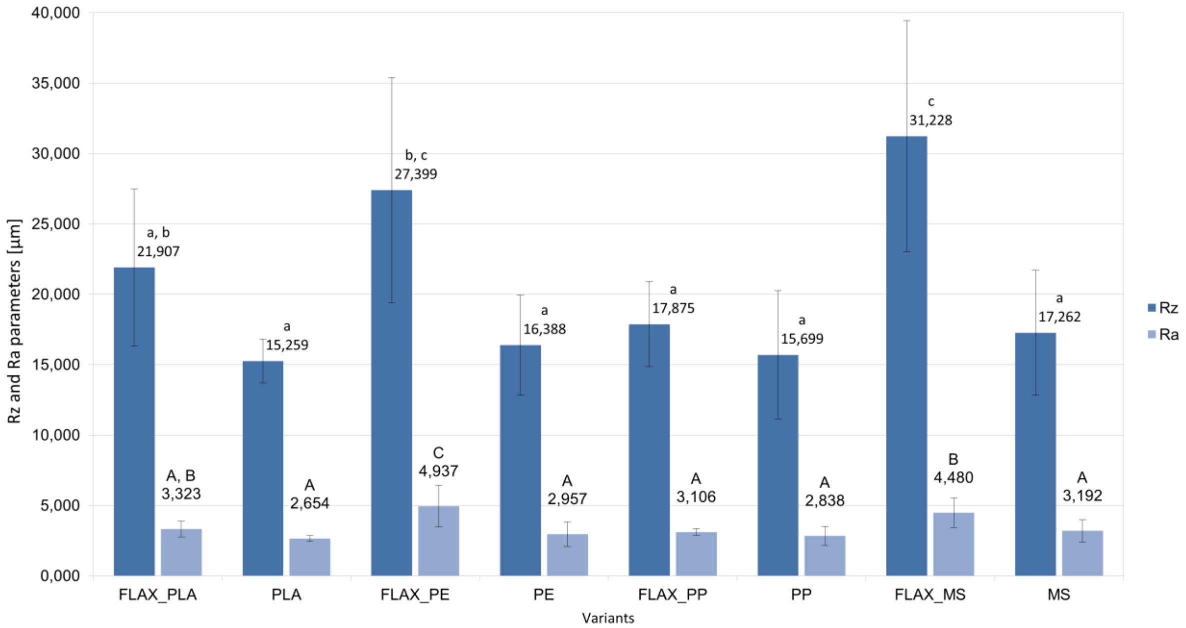

Figure 10. Surface roughness parameters for composite variants and control samples (pure polymers: MS, PE, PP, PLA); a, b, c A, B,C – homogeneous groups by Tukey test (α = 0.05)

CONCLUSION

FLAX_PP composite were characterized by higher mechanical parameters than other composites. FLAX_PP also occured to be the most resilient composite with the similar properties to pure polypropylene. FLAX_PP and FLAX_PE composites are resistant to 24-hour water exposure - they swell, but their structure is not destroyed. FLAX_PLA samples absorbed a lot of water and partially disintegrated. The reason for this may be their increased porosity. FLAX_MS composite is not resistant to water - it broke down as early as 2h after immersion. All tested composites are hydrophilic in nature. All composites tested are medium rough materials. Further researches should be conducted to improve quality parameters of designed composites. Alternative lignocellulosic raw materials from agri-food production could be good alternative for wood in composite production.

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Streszczenie: *Wytłoki lniane jako substytut surowca drzewnego w technologii kompozytów lingocelulozowych*. Kompozyty wykonane z wytłoków z nasion lnu oraz HDPE, PP, PLA i skrobi poddano testom parametrów jakościowych. Zbadano gęstość, wytrzymałość na zginanie i moduł sprężystości, a także nasiąkliwość, zwilżalność i chropowatość powierzchni. Dodatek lnu do różnych polimerów spowodował znaczące zmiany parametrów, ale poziom wpływu dodatku różnił się między polimerami. Chociaż wykazano, że parametry jakościowe wytworzonych biokompozytów są niższe od czystych polimerów, należy podjąć wysiłki w celu poprawy tych właściwości. Produkty uboczne z przetwórstwa rolno-spożywczego mogą być cennym surowcem do produkcji nowych przyjaznych dla środowiska materiałów.

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