

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

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Abstract: *Corn pomace as a substitute for wood raw material in lignocellulosic composite technology.* Composites made from polymers PE, PP, PLA and modified starch with addition of corn pomace were produced and subjected quality parameters test. There were examined such parameters as: density, static bending strength and modulus of elasticity and also water absorption and thickness swelling after 2 h and 24 h soaking in water, wettability and surface roughness. In general, the addition of corn pomace to polymers caused changes of parameters and through comparing parameters of different composites and control samples it was possible to indicate corn composite variants that occurs to have similar properties to pure polymer samples. Waste products should be seriously considered as a valuable substitute of wood raw material, but characteristics of composites should be improved through further research.

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

INTRODUCTION

Wood based products as materials formed from solid wood or other lignocellulosic material, as a result of shredding and recombining, are widely used in: furniture, construction, transportation equipment, packaging production, among others (Styczynski 2006). In recent years, with the rapid development of the timber industry, attention has been increasingly drawn to potential future problems in the availability of wood raw material for industrial use. This situation is related, among other things, to the European Union's zero-emission policy, which aims to reduce greenhouse gases by at least 55 % by 2030 and achieve climate neutrality in 2050 (European Commission 2021). One of the measures to achieve the targets is to reduce timber harvesting by up to 40 % in EU member states, which could have a negative impact on the furniture industry, which is especially important for Poland.

According to data for 2022, the production of particleboard (for use mainly in furniture making) in Poland reached nearly 5 million m³, which is 4.74 % of global production, that makes Poland the world's fifth largest producer of this type of wood products (<https://www.fao.org/faostat/en/#data/FO>). The production and export of furniture in Poland already in 2013 accounted for about 2.48 % of global production (Ulbrych 2016), and according to data for 2021 it increased to 5.4 % of global production (Department of Economic Analysis PKO Bank Polski S.A. 2022). The search for an alternative raw material to wood lignocellulosic fibers is economically justified due to the fact that the furniture sector accounts for 2.4 % of Poland's GDP .

Another important economic aspect is the use of post-production by-products. Poland, as a member state of the European Union, has pledged in 2019 to move towards a circular economy and comply with European recommendations on state environmental policy. Realization of the set goals is related to, among other things, reusing tens of millions of tons of post-consumer plastics and designing materials so that they can be easily recycled in the future or significantly extend the life cycle of products made with them (Kulczycka 2019).

One of the more interesting materials, the production of which is based on combining post-consumer (often waste) products, are WPC (wood plastic composites), which could feasibly affect less wood consumption and better use of post-consumer polymers. Polymer-wood composites are formed by combining two basic components, which are generally thermoplastic polymers and wood particles or other fibers of plant origin (Borysiuk 2012). The material specificity of the production of WPC allows the use of alternative raw materials. On the one hand, it is possible to use post-consumer polymers, and on the other, lignocellulosic particles obtained from raw materials other than wood, often from the agri-food industry.

Potential raw materials for obtaining lignocellulosic fibers can be production residues from agri-food processing such as: stalks of most cereals, coconut fibers, peanut shells, corn cobs (Nourbakhsk and Ashori 2009) or for example, perennial grasses i.e. *Miscanthus* (*Miscanthus giganteus* J. M. Greef & Deuter ex Hodk. & Renvoize)) (Frąckowiak et al. 2008). Alternative raw materials to wood have been studied, mainly in terms of their usefulness in the production of lignocellulosic plastics of various types - one of reviewed raw materials is sunflower husk. Through research, it has been proven that sunflower husk is suitable for the production of particleboard not dedicated to construction purpose, but the use of this type of raw material significantly deteriorates the mechanical and physical properties of the resulting board (Borysiuk et al. 2020).

Data compiled by the National Center for Agricultural Support (NEB), which analyzed the share of individual cereals in sowing in the Polish area, shows that the second largest share, right after wheat, is in the corn crop. During the process of harvesting the grains, they are mechanically separated from the stalk and leaves (Zhangfeng et al. 2016). Some of the grain thus extracted goes as an intermediate product for sale, and some goes for further processing into corn oil or corn starch, among others, during which a by-product called bagasse or corn pomace is produced. This is the skimmed part of the corn kernel, which so far has not found widespread use in industry beyond the production of livestock feed in the form of dried stock or as a meal for fishing groundbait (Strzetelski 2006). In the present study, the possibility of using corn seeds pomace as a by-product of oilseed crop processing in polymer-lignocellulose composite technology was determined for four different polymers used as a binder, i.e. polypropylene (PP), polyethylene (PE), polylactide (PLA) and modified starch (MS).

MATERIALS

Within the framework of the work, lignocellulose-polymer composites were produced from a combination of corn post-oil pomace (AL-PHADAR, Nienaszow, Poland) and four types of thermoplastic polymers. Four material variants differing in the polymer used were made: corn-polyethylene (CORN_PE) HDPE (Orlen Polyolefins Sp. z.o.o., Plock, Poland), corn-polylactide (CORN_PLA) (Resinex Poland Sp. z.o.o., Warsaw, Poland), corn-polypropylene (CORN_PP) (ORIGIN) and corn-modified starch (CORN_MS) (envifill® GP, Grupa Azoty S.A., Tarnów, Poland). The boards were manufactured for an assumed density of 900 kg/m³ and a thickness of 2.5 mm.

The manufacture of pellets for the individual blends required milling the corn pomace into a fine fraction, the dimensions of which did not exceed 300µm, using a DS-2/OBR chip cutter (OB-RPPD Sp. z o.o., Czarna Woda, Poland). After such preparation, the material was mixed with polymers at a weight ratio of 50/50. The next step was to extrude the composite using an extruder (Leistritz Extrusionstechnik GmbH, Nürnberg, Germany). The temperature during

extrusion varied between 170-180 °C, depending on the section of the machine. The granules thus formed were set aside to cool, and then ground in a DS-2/OBR chip cutter (OB-RPPD Sp. z o.o., Czarna Woda, Poland). The granulate manufacturing process was carried out at the Research and Development Center of the Wood-based Panel Industry Sp. z o.o. in Czarna Woda.

The next stage involved the manufacture of boards from the resulting granulate, using a flat-press process in a shelf press. After the granules were manually distributed in the previously prepared frame, material was subjected to a pressing process with the following pressure parameters: a pre-pressing stage of 0.05 MPa for 45 seconds, calculated from the contact of the press shelves with the carpets, a proper pressing stage of 0.5 MPa for 75 seconds, and a final pressing stage of 1.0 MPa for 180 seconds. Subsequently, the samples were cooled for about 10 minutes in a shelf press (Industrial Equipment Plant, Nysa, Poland) at 1.0 MPa. The variable parameter during pressing, depending on the polymer used, was the shelf temperature of the used press (AB AK Eriksson, Mariannelund, Sweden). Table 1 presents pressing temperature for corn-polymer composites and control samples (pure polymers: MS, PE, PP, PLA).

Table 1. Pressing temperature for corn-polymer composites and control samples (pure polymers: MS, PE, PP, PLA)

Variant	Temperature [°C]
CORN_PLA	200
PLA	180
CORN_PE	200
PE	140
CORN_PP	180
PP	180
CORN_MS	160
MS	180

In the case of the corn-starch mixture, water was added to achieve the proper consistency of the pressed boards. In order to determine its optimal amount, 6 boards were produced with an additional proportion of water of 9 %, 12 % and 15 %, relative to the weight of the prepared corn starch mixture. These boards were pressed at 140 °C and 160 °C with the above-described pressing procedure and cooling as for full-size slabs. After making (as a pilot study) the boards and performing an organoleptic evaluation, it was concluded that the 9 % water added to the components relative to the weight of the mixture and the temperature of 160 °C were the optimal parameters.

To determine selected physical and mechanical properties of manufactured composites and control samples of pure polymers, they were subjected number of tests. Density was defined according to PN-EN 323:1999 - Wood-based panels - Determination of density. Static bending strength and modulus of elasticity according to PN-EN 310:1994 - Wood-based panels - Determination of bending modulus and bending strength. The tests were conducted using a strength apparatus manufactured by OB-RPPD Sp. z o.o., Czarna Woda, Poland and using OBRCzW_NET_MS software. Thickness swelling (TS) after 2 h and 24 h soaking in water were determined 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 expressed in % 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, $n \in (2, 24)$. The surface contact angle test was performed using a Haas Phoenix 300 goniometer (Surface Electro Optics, Suwon City, South Korea). After placing a drop of

distilled water on the surface of the test sample, images were taken at 5, 20, 40 and 60 seconds after the drop was placed, and with Image XP v. 5.8 image analysis software (Surface Electro Optics, Suwon City, South Korea), average contact angle and its change in time for each sample was determined. Using SurfTest SJ-210 portable contact profilometer (Mitutoyo Co., Kawasaki, Japan), the surface roughness parameters were defined. The following amplitude parameters were determined: Rz, Rq and Ra. Tests were conducted according to 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.

Statistical analysis were performed using Statistica version 13.3. It was performed for the significance level $\alpha = 0.05$.

RESULTS

After performing the planned tests, the results obtained were analyzed, taking into account the effect of the filler on the individual polymers. Table 2 shows the results of the average densities of the tested samples. Analyzing the data, we can observe a slight increase in the density of samples containing the addition of corn, compared to the control samples of pure polymers. Only in case of composite with MS we observed the opposite effect. The same observation occurs while analysis of density test results.

Table 2. Thickness of corn-polymer composites and control samples (pure polymers: MS, PE, PP, PLA)

Variant	thickness [mm]	standard deviation [mm]	coefficient of variation [%]
CORN_PLA	2.16	0.07	3.13
PLA	2.22	0.07	3.07
CORN_PE	2.33	0.08	3.29
PE	2.58	0.03	1.32
CORN_PP	2.34	0.06	2.57
PP	2.64	0.10	3.84
CORN_MS	2.42	0.06	2.60
MS	2.13	0.02	1.11

Table 3. Density of corn-polymer components and control samples (pure polymers: MS, PE, PP, PLA)

Variant	density [kg/m ³]	standard deviation [kg/m ³]	coefficient of variation [%]
CONR_PLA	1070	34.95	3.27
PLA	1061	23.88	2.25
CORN_PE	980	26.65	2.72
PE	897	5.05	0.56
CORN_PP	1019	17.60	1.73
PP	873	18.33	2.10
CORN_MS	967	49.07	5.07
MS	1062	2.22	0.21

After examining the static bending strength and presenting the results in Figure 1 it can be observed that the corn extrudate reduced the flexural strength of the polymers. This is most pronounced in the case of the variant with polylactide. The smallest decrease was recorded for polyethylene, where the difference between the control sample and the corn-polyethylene

sample was not significant according to homogenous groups. Pure MS sample was not stiff enough to conduct a static bending strength test. Addition of corn particles resulted in sufficient stiffness, and it was possible to test the MS_CORN sample. As it can be observed CORN_MS and CORN_PE are in the same homogeneous group as pure PE.

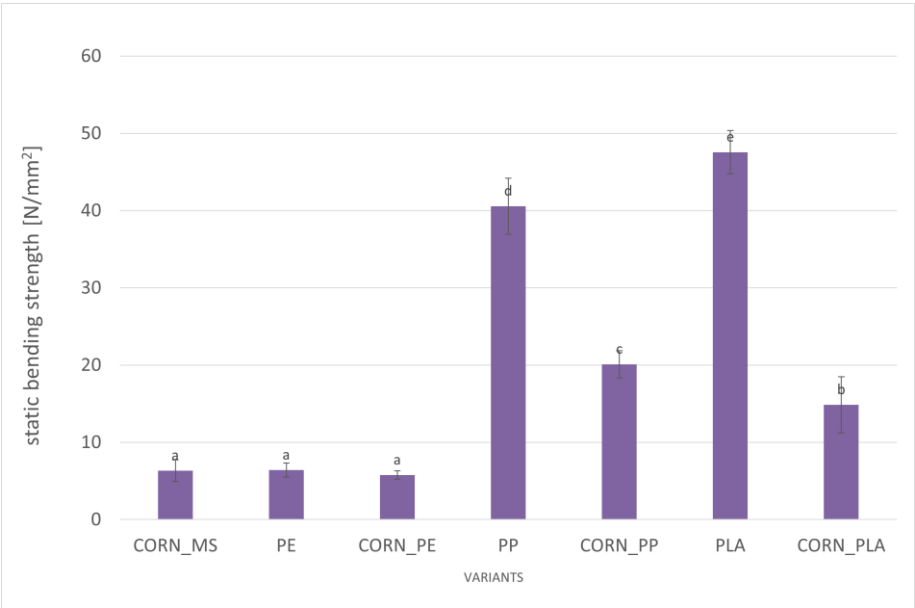


Figure 1. Static bending strength for corn-polymer composites and control samples (pure polymers: MS, PE, PP, PLA); a, b, c, d, e – homogeneous groups by Tukey test ($\alpha = 0.05$)

An equally important parameter is the deformability of the tested plates. Figure 2 presents the average modulus of elasticity for composites and control samples. The addition of corn pomace to the polymers lowered their modulus, only in the case of the corn-polyethylene the difference was not significant according to homogeneous groups. The decrease effect in the value of modulus of elasticity, after addition of corn, was similar for variants with PLA and PE. Variant CORN_MS showed a higher modulus than CORN_PE and PE.

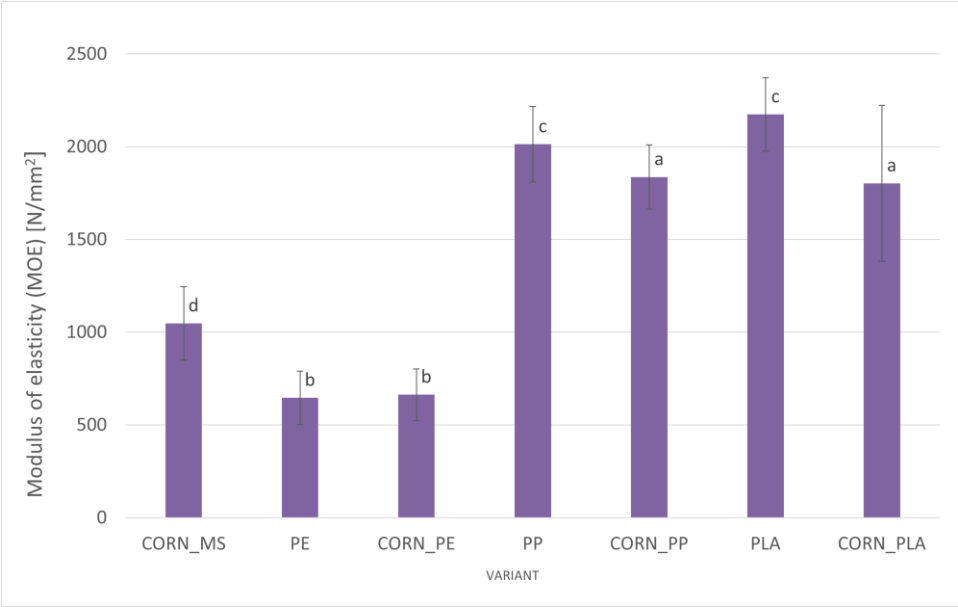


Figure 2. Modulus of elasticity for corn-polymer composites and control samples (pure polymers: MS, PE, PP, PLA); a, b, c, d – homogeneous groups by Tukey test ($\alpha = 0.05$)

In the case of furniture boards, an equally important quality parameter is surface wettability, which shows the interaction of the tested surface with liquids, mainly water (Kuszelnicka et al. 2018). Indicating homogenous groups for each time it was possible to observe that for all pairs of polymer and composite there were no significant differences except MS and CORN_MS. Addition of corn in case of MS and CORN_MS samples caused a significant increase of contact angle. Results are presented in Table 4.

Table 4. Contact angle values measured 5, 20, 40 and 60 seconds after droplet application; a, b, c, d, e – homogeneous groups by Tukey test ($\alpha = 0.05$) determined separately for particular time

Time	5s		20s		40s		60s	
Variant	contact angle [°]		contact angle [°]		contact angle [°]		contact angle [°]	
MS	55.970	e ₅	44.883	e ₂₀	39.941	d ₄₀	37.316	d ₆₀
CORN_MS	85.760	d ₅	83.352	d ₂₀	80.124	a ₄₀	77.701	a ₆₀
PE	70.331	a ₅ , b ₅ , c ₅	69.722	a ₂₀ , b ₂₀ , c ₂₀	69.048	a ₄₀ , b ₄₀ , c ₄₀	68.226	a ₆₀ , b ₆₀ , c ₆₀
CORN_PE	73.086	a ₅ , b ₅ , c ₅	71.818	a ₂₀ , b ₂₀ , c ₂₀ , d ₂₀	71.143	a ₄₀ , b ₄₀ , c ₄₀	69.999	a ₆₀ , b ₆₀ , c ₆₀
PP	76.954	b ₅ , c ₅ , d ₅	76.821	b ₂₀ , c ₂₀ , d ₂₀	75.994	a ₄₀ , c ₄₀	75.523	a ₆₀ , c ₆₀
CORN_PP	81.512	c ₅ , d ₅	81.200	c ₂₀ , d ₂₀	80.443	a ₄₀	79.448	a ₆₀
PLA	62.625	a ₅ , e ₅	61.064	a ₂₀	60.005	b ₄₀	59.740	b ₆₀
CORN_PLA	69.298	a ₅ , b ₅	67.671	a ₂₀ , b ₂₀	66.342	b ₄₀ , c ₄₀	65.549	b ₆₀ , c ₆₀

One of the last composite parameters tested was surface roughness. To define surface properties, array of characteristics like Rz, Ra and Rq was determined. The most sensitive for local surface changes is Rz parameter. Rz, Ra and Rq parameter values for CORN_MS occurred to be significantly higher than others.

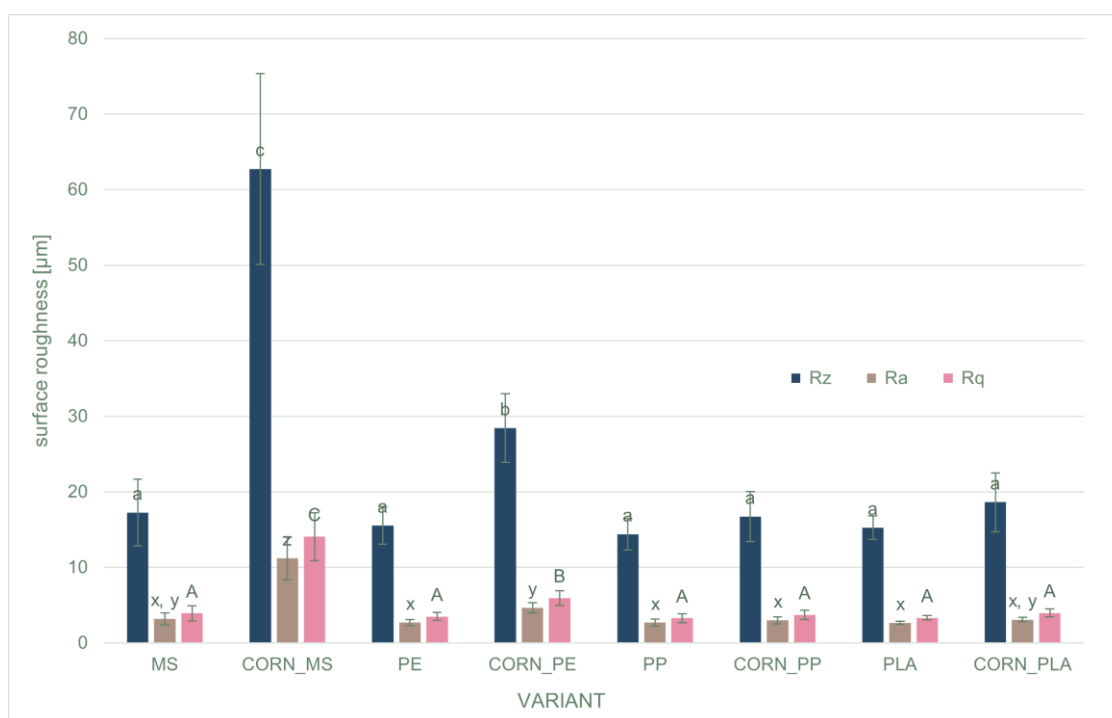


Figure 3. Rz, Ra and Rq parameters for corn-polymer composites and control samples (pure polymers: MS, PE, PP, PLA); a, b, c, x, y, z, A, B, C – homogeneous groups by Tukey test ($\alpha = 0.05$)

For all parameters MS, PE, PP, CORN_PP, PLA and CORN_PLA samples were included in the same homogenous groups.

The last two tested parameters are the water absorption and thickness swelling after soaking in water after 2 h and 24 h. The results are shown in Figures 5 and 6. It can be observed that the addition of corn pomace increased the absorbency and swelling of the tested samples, thereby enhancing the hygroscopicity. In the case of absorbability, it can be observed that the highest value after 24 h soaking in water for the corn-poly lactide samples, while the lowest in corn-polypropylene samples.

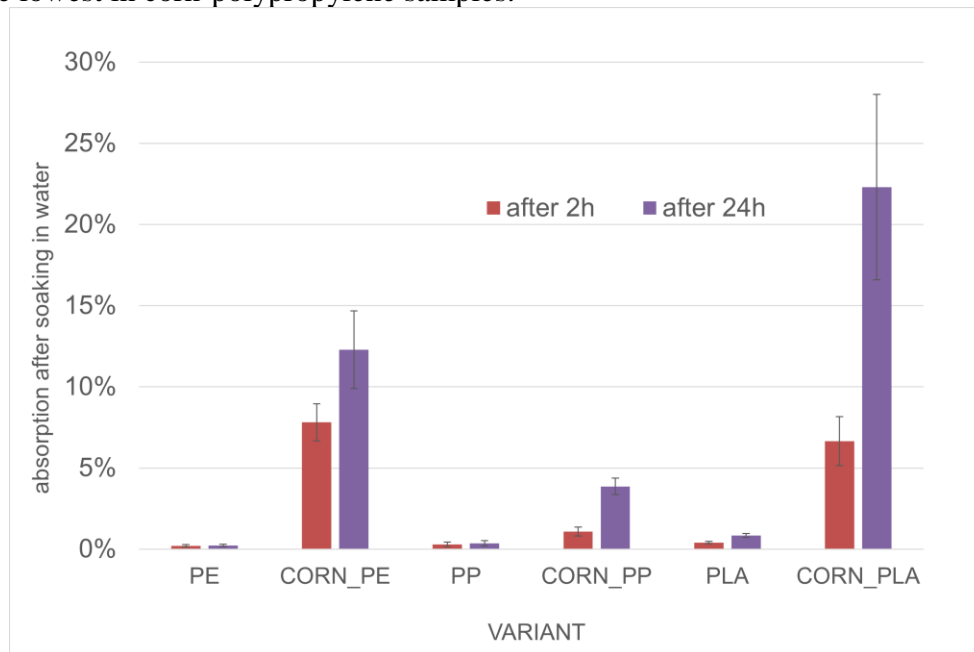


Figure 4. Absorption after soaking in water of corn-polymer composites and control samples (pure polymers: MS, PE, PP, PLA)

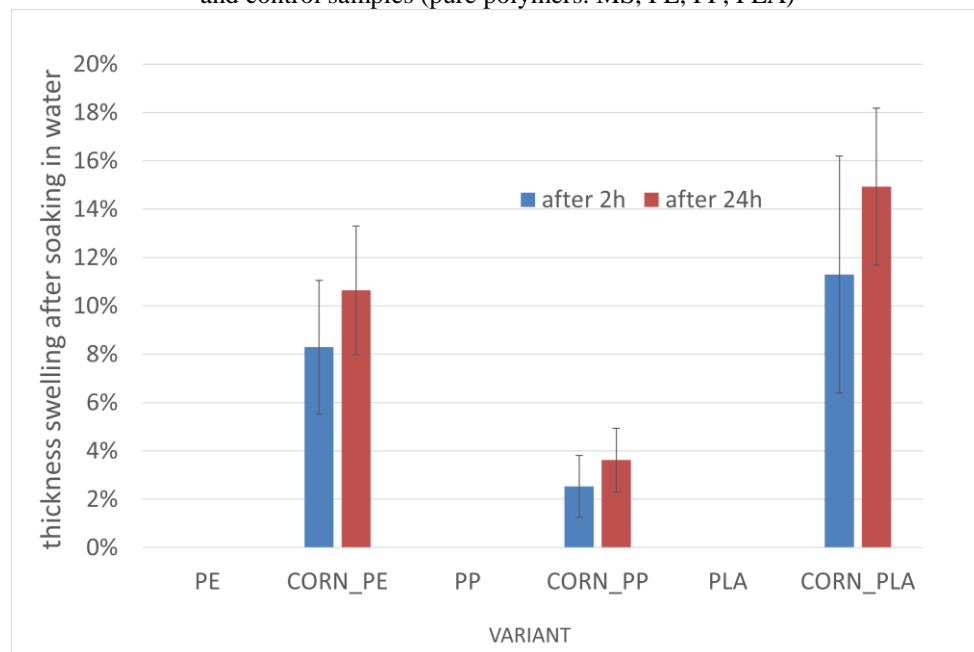


Figure 5. Thickness swelling after soaking in water of corn-polymer composites and control samples (pure polymers: MS, PE, PP, PLA)

Control samples made of pure polymer were characterized by close to complete hydrophobicity. Similar behavior of the samples can be seen in the case of swelling. The

largest value this time is characterized by the corn-polyethylene variant, the smallest, as in the case of swelling, by corn-polypropylene. The swelling of the polymers is close to 0 %.

CONCLUSION

The use of by-products from agri-food processing offers many opportunities for further development of polymer-lignocellulosic composites technology, but from the vastness of available raw materials it would be necessary to select those giving the possibility of producing composites with similar quality parameters to those created using wood raw material. Studying composites based on polymers and lignocellulosic particles derived from corn pomace it was possible to obtain composites that can certainly compete with wood based materials in certain spheres. However, in order to be able to fully replace wood, the whole process of producing composites using corn and thermoplastic polymers needs to be improved, so that the whole process is efficient and reproducible. The resulting composites could be used in the production of furniture components, decorative wall panels or packaging materials. Any research conducted in the future should focus, but not only, on improving component structure and mechanical properties. Equally important is the optimization of the proportions of mixtures and the creation of one most efficient, which will combine the best quality and economic parameters.

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Streszczenie: *Wytłoki kukurydziane jako substytut surowca drzewnego w technologii kompozytów lingo celulozowych.* Wytworzono kompozyty z polimerów HDPE, PP, PLA oraz skrobi modyfikowanej z dodatkiem wytłoków ziaren kukurydzianych i poddano je badaniom parametrów jakościowych. Zbadano takie parametry jak: gęstość, wytrzymałość na zginanie statyczne i moduł sprężystości, a także spęcznienie na grubość oraz nasiąkliwość po 2 i 24 godzinach moczenia w wodzie, zwilżalność i chropowatość powierzchni. Dodatek wytłoków kukurydzianych do polimerów spowodował zmiany parametrów, a poprzez porównanie parametrów różnych kompozytów i próbek kontrolnych możliwe było wskazanie wariantów kompozytów zawierających wytłoki kukurydziane, które mają podobne właściwości do próbek czystego polimeru. Pozostałości produkcyjne przemysłu rolno-spożywczego powinny być brane pod uwagę jako wartościowy substytut surowca drzewnego, jednak parametry ich wytwarzania powinny być zoptymalizowane czemu służyć będą dalsze badania.

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