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Characteristics of high-density fibreboard produced with the use of rice starch as a binder

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Abstract: *Characteristics of high-density fibreboard produced with the use of rice starch as a binder*. The adhesives used as binders in the boards are not biodegradable, and their formaldehyde is toxic. Nowadays it is very important to take care of the environment. There are a lot of biodegradable products that as well as the current board binders can meet good mechanical and physical properties and at the same time not harm the environment. Therefore in this project, the study aimed to investigate the possibility of producing dry-formed fibreboards using rice starch as a binder. The research involved the production of boards with a mass proportion of rice starch 0%, 10%, 12%, 15%, and 20% (when referred to as totally dry wood fibre mass) and to study of their selected physical and mechanical properties. The results proved that rice starch can be used as a binding agent if we choose the right amount of starch so that it can improve some mechanical and physical properties. Very good properties came out for the determination of modulus of elasticity in bending and of bending strength and for screw withdrawal resistance with a high rice binder content, but on the other hand for internal bonds, the high rice content reduced the properties. For some studies, rice flour improved properties but not enough to comply with standards as was the case with the swelling of the thickness.

Keywords: fibreboard, HDF, rice starch, binder.

INTRODUCTION

Nowadays, a lot of waste is generated, which is very bad for the environment. The adhesives used as binders in the wood-based composites production are non-biodegradable and the formaldehyde in them is toxic. Formaldehyde in higher concentrations is classified as a possible human carcinogen (Chrobak et al. 2022). Urea-formaldehyde (UF), melamine-formaldehyde (MF), phenol-formaldehyde (PF), and other combinations of amine resins are the most often used in the current production of adhesives for wood-based panels (Huang et al. 2022). High-density fibreboards (HDF) are made from wood fibres and formaldehyde-based resin (mostly UF or MUF). That's why nowadays scientists are focusing on the search for bio-based adhesives because the adhesives used for wood are synthesized from non-renewable and toxic resources making them non-recyclable. Starch is a natural polymer with the distinction of being biodegradable, renewable, and comparably inexpensive. Thanks to this, it shows greater potential as an alternative adhesive for HDF boards (Adly et al. 2020). Starches are mainly extracted from the roots, stems, and seeds of staple crops such as rice, corn, wheat, tapioca, and potato (Gadhave et al. 2017). Yearly starch production from cereals is about 2050 million tons and from roots and tubers about 679 million tons (Tester. R.F and Karalas. J, 2002). Starch is the main source of energy in the human diet but it is also used to glue paper, as an additive in cement or as an additive in gypsum plaster and gypsum-fiber board (Burrell 2003).

Wronka et al. (2020) have researched the use of potato starch in boards made wet using different starch contents. In the study, boards with 0, 1, 5, 10 and 20% starch content were made. The results showed that as the starch content of the board increases, the value of the mechanical properties also increases. Variant 20 shows the highest elastic modulus value of 3900 N/mm^2 and is about 27% higher than the reference sample which is 3077 N/mm^2. And, the results for the rupture modulus for variant 20 came out about 138% higher compared to the reference sample.

Starch was tested in medium-density fibreboard (MDF) where oxidized starch (OS) was used as a binder (Adly et al. 2020). To adjust the performance of OS adhesives, they used four molar ratios of H_2O_2/starch, three content levels of crosslinking agents, and two types of crosslinking agents, i.e. blocked polymeric methyl diphenyl diisocyanate (B-PMDI) and citric acid (CA). The results showed that the use of oxidized starch as a binder in MDF technology showed good physical and mechanical properties.

Rosa and Kowaluk (2022) have studied the effect of plant glue as a binding agent in MDF. They created five types of boards using different proportions of mass plant glue. The results during the modulus of rupture (MOR) test show that the higher the proportion of plant glue, the better the results, in this case, a resination of 20% leads to the highest MOR value, 42.9 N/mm^2. Similar results came out during the modulus of elasticity (MOE) test where the highest result came out for a 20% mass share of the adhesive and the value was 2883 N/mm^2.

Sulaiman et al. (2013) studied the experimental panels' physical and mechanical properties, which were made of rubber wood particles and oil palm starch modified with epichlorohydrin as a binder. The panels were created with a density of 0.60 g/cm^3 and 0.80 g/cm^3 at two pressing times of 15 and 20 min. The results for panels with a density of 0.80 g/cm^3 produced using modified oil palm starch and a pressing time of 15 min showed better properties than the other boards. The smallest thickness swelling values were found in panels with a density of 0.60 g/cm^3 at a pressing time of 15 min. Based on the results of the study, it can be concluded that epichlorohydrin-modified oil palm starch can be used as an environmentally friendly binder.

Interesting studies have been conducted on obtaining biocomposites of manioc starch with eucalyptus wood particles using thermal compression (Lomelí-Ramírez et al. 2014). The composites that were obtained varied in wood particle content from 5 to 30% and in size from 4 to 8 mm. The tensile strength and elastic modulus of the biocomposites increased with the addition of eucalyptus particles from 5 to 30%. Biocomposites created with smaller particle sizes (4 mm) showed better tensile properties thanks to better adhesion between matrix and particles. Moisture absorption results decreased inversely proportional to the amount of wood particles contained in the matrix. The effect of particle size in biocomposites with similar particle content was not significant.

This study aimed to investigate the selected mechanical and physical properties of HDF boards produced with different proportions of rice starch used as binder. In the scope of the study, high-density fibreboards with different amounts (w/w) of rice starch binder (0%, 10%, 12%, 15%, and 20%) were created in laboratory conditions and selected mechanical and physical properties of the produced boards were investigated. There is a lot of research related to starch as an additive in glueing various boards, and various possibilities of using starch in wood technology or its modification, but the use of rice starch in HDF hardboard as a binder has not been investigated so far.

MATERIALS AND METHODS

Materials used to create the test samples:

- Rice starch in the form of flour (MELVITA S.A. 02-390 Warszawa, Grójecka St. 194/91)
- Distilled water
- Industrial fibres made from 95% pine (*Pinus sylvestris* L.) and spruce wood (*Picea abies* (L.) H.Karst) by weight with a moisture content (MC) of about 3%
- Urea-formaldehyde (UF) resin Silekol S-123 (Silekol Sp. z o. o., Kędzierzyn-Koźle, Poland) as a reference binder; molar ratio of 0.9, and solid content of 66.5% has been used. The resination was set at 12% dry resin calculated on dry fibres with 3.0% of ammonium nitrate hardener, both calculated regarding dry resin content. The curing time of the adhesive mass composed as mentioned above at 100 °C was about 82 s. No further hydrophobic agents have been added.

The fibres were placed in a drum blender, and after activation of the machine, rice starch was sifted through a sieve, and water was sprayed on the fibres using an air gun. The amount of water was set for every panel variant, depending on the rice binder content. The water/rice flour ratio was constant and it was 0.5 w/w. The mixed fibres were formed into mats and prepared for pressing. They were hot pressed (hydraulic press AKE, Mariannelund, Sweden) at 200 °C for 20 s/mm of nominal panel thickness (3 mm), assumed nominal density 900 kg/m^3 and maximum unit pressure of 2.5 MPa. The maximum press pressure was kept for the initial 50% of the pressing time, then was reduced by 1/3 for the next 20% of the pressing time, then again reduced by 1/3 for the next 20% of the pressing time, and after this, the pressure was continuously reduced to open the press within the remaining 10% of the total pressing time. The produced boards were conditioned before the tests at 20 °C and 65% ambient air humidity until a constant mass had been obtained.

Depending on the amount of starch used (w/w), panels were created in five variants:

1. Fiberboard with 10% rice starch as a binder (hereafter called R10)
2. Fiberboard with 12% rice starch as a binder (hereafter called R12)
3. Fiberboard with 15% rice starch as a binder (hereafter called R15)
4. Fiberboard with 20% rice starch as a binder (hereafter called R20)
5. Fiberboard with urea–formaldehyde (reference; hereafter called REF)

Determination of Modulus of Elasticity in Bending and of Bending Strength

The elasticity and strength in bending were performed according to EN 310 (1993) on a computer-controlled universal testing machine on at least 8 samples in each variant.

Tensile strength

Tensile strength is perpendicular to the plane of the panel (Internal bond – IB) and was tested according to EN 319 (1993) on a computer-controlled universal testing machine on at least 8 samples in each variant.

Screw withdrawal resistance

Screw withdrawal resistance (SWR) was tested according to EN 320 (2011) on a computer-controlled universal testing machine on at least 8 samples in each variant.

All the mechanical properties were examined with a computer-controlled universal testing machine delivered by the Research and Development Centre for Wood-Based Panels Sp. z o.o. (Czarna Woda, Poland).

Contact angle

The contact angle was taken on a PHOENIX 300 goniometer (SEO Co. Ltd., South Korea), where the samples were wiped with a drop of water. The test was repeated 3 times on each variant. Images and results after 0 and 60 seconds of drop deposition were used for the study.

Density profile

The density profile of samples was analyzed using a DA-X measuring instrument (Fagus-GreCon Greten GmbH and Co. KG, Alfeld/Hannover, Germany). The measurement based on direct scanning X-ray densitometry was carried out with a speed of 0.05 mm/s across the panel thickness with a sampling step of 0.02 mm. Samples were cut into 50 mm × 50 mm nominal dimensions. Three samples of each type of composite were used to study the density profile.

Water absorption (WA), swelling in thickness (TS)

The water absorption and thickness swelling were tested according to EN 317 (1993) after 2 hours and after 24 hours of soaking in water on at least 8 samples in each variant.

Surface water absorption

The surface absorption was tested according to EN 382-2 (1993) after 2 hours on 2 samples from each variant.

Statistical Analysis

One-way analysis of variance (ANOVA) was conducted to study the statistical significance of the differences in mean values of the above-mentioned parameters on the properties of the tested panels at the 0.05 significance level ($P = 0.05$). All the statistical analyses were performed using the software IBM SPSS Statistics 22. The statistically significant differences in achieved results, whenever the data were evaluated, are given in the Results and Discussion section.

RESULTS AND DISCUSSION

Determination of Modulus of Elasticity in Bending and of Bending Strength

The results for the modulus of elasticity and the modulus of rupture in static bending are shown in Figure 1. The graph shows that the modulus of elasticity of each tested sample with rice binder is higher than that required by EN 622-5 (2010). As the addition of rice glue increases, the modulus of elasticity increases. The lowest MOR value came out for sample R10 and was 24.8 N/mm^2; the highest MOR values are presented by sample R20 of 40.2 N/mm^2. As can be seen from the chart, no sample with rice binder for MOR has higher values than the reference sample whose value is 42.2 N/mm^2. The modulus of rupture for rice binder The R20 sample has a higher MOE value than the reference sample whose value is 3677 N/mm^2. The MOE value for all samples with rice binder is higher than that required by EN 622-5 (2010). In the case of MOR and MOE, the only statistically significant difference has been found for 20% resination when referring to the remaining panels with rice binder.

Similar results for the increase in MOR and MOE came out when studying hardboard panels bonded with potato starch (Wronka et al. 2020) and when studying citric acid-modified corn starch used as a binder for wood composites (Hazim et al. 2020). In these studies, MOR and MOE values increased with increasing starch addition.

In the research of Sulaiman et al. (2013) mechanical properties showed up highest for panels bound with modified starch. The highest average MOR and MOE values for the higher panel density (0.80 g/cm^3) were 19.09 N/mm^2 and 3471.64 N/mm^2, accordingly.

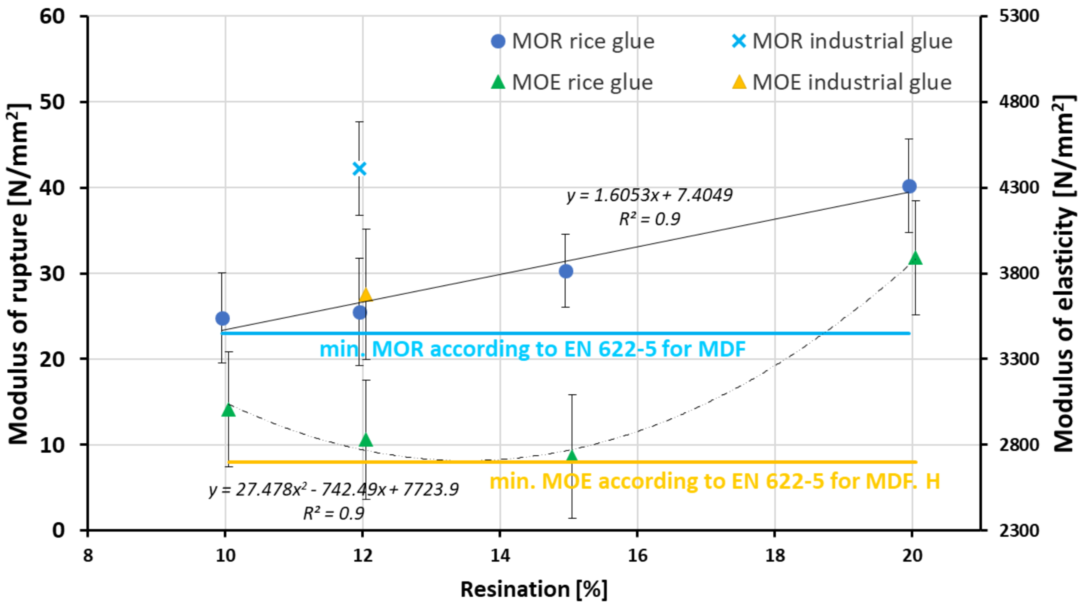

Figure 1. Modulus of rupture and modulus of elasticity of the boards of various content of rice binder

Internal bond

The results for the internal bonding of the manufactured rice-bonded panels are shown in Figure 2. The lowest IB result for rice glue samples came out for sample R20 of 0.13 N/mm^2, and the highest result came out for samples R10 and R12 of 0.27 N/mm^2. IB for the reference sample glued with UF glue was 0.72 N/mm^2. In the chart, we can see that all samples glued with rice glue have lower values than the requirements of EN 622-5 (2010). The greater the addition of rice glue, the lower the results. The statistically significant differences in average

IB have been found for panels with 15% rice flour resination when referred to panel R12, as well as the statistically significant difference has been found between panels R12 and R20.

Sulaiman et al. (2013) studied particleboards made from oil palm starch modified with epichlorohydrin where the results for IB show the best mechanical properties for boards with modified starch. Similar results came out during a study on starch modified with citric acid in the production of wood composites (Hazim et al. 2020) where all tested samples of the wood composite satisfied the IB requirements according to the specified standard. In contrast, research by Theng et al. (2017) showed lower IB properties for fibreboard with lignin added.

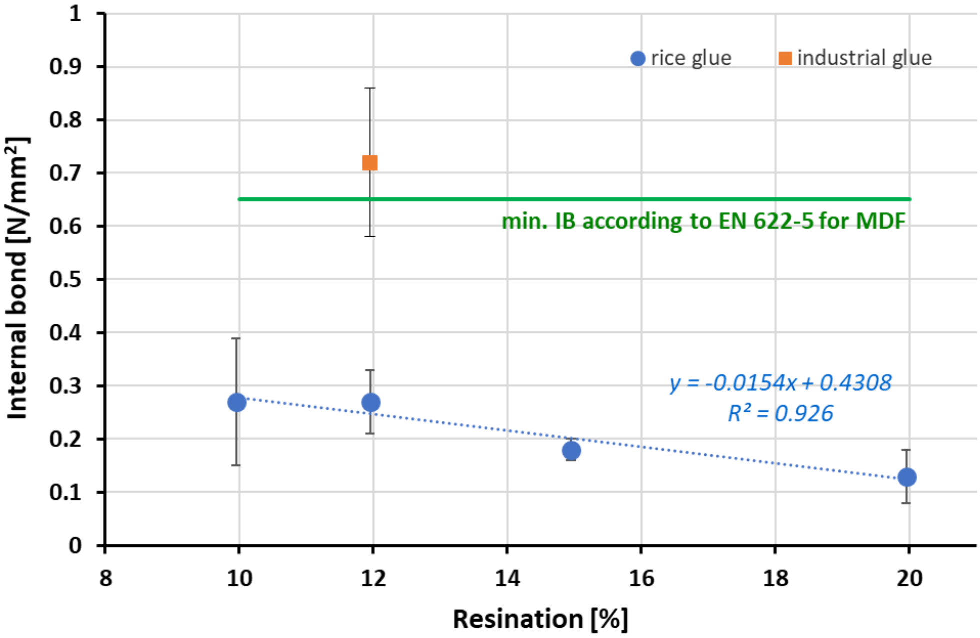

Figure 2. The internal bond of the boards of various content of rice binder

Screw withdrawal resistance (SWR)

The results for the screw drawing from panels with different rice binder contents are shown in Figure 3. The lowest SWR results came out for the R10 and R15 rice-bonded samples of 124 N/mm, and the highest results came out for the R20 sample of 145 N/mm, which is higher than the reference sample of 141 N/mm. The graph shows that a properly selected rice glue amount in the test samples can give better properties than reference samples. The only statistically significant difference between SWR average values has been found for 20% resination panels when referring to the remaining panels with rice binder.

Rosa and Kowaluk (2022) also obtained results showing that an increase in resination leads to a significant increase in SWR when testing medium-density fiberboard bonded with plant glue. The lowest results came out for the variant with the smallest amount of plant binder 116 N/mm, and the highest results came out for the variant with the highest amount of plant binder 144 N/mm. However, no sample with plant glue had higher values than the reference sample

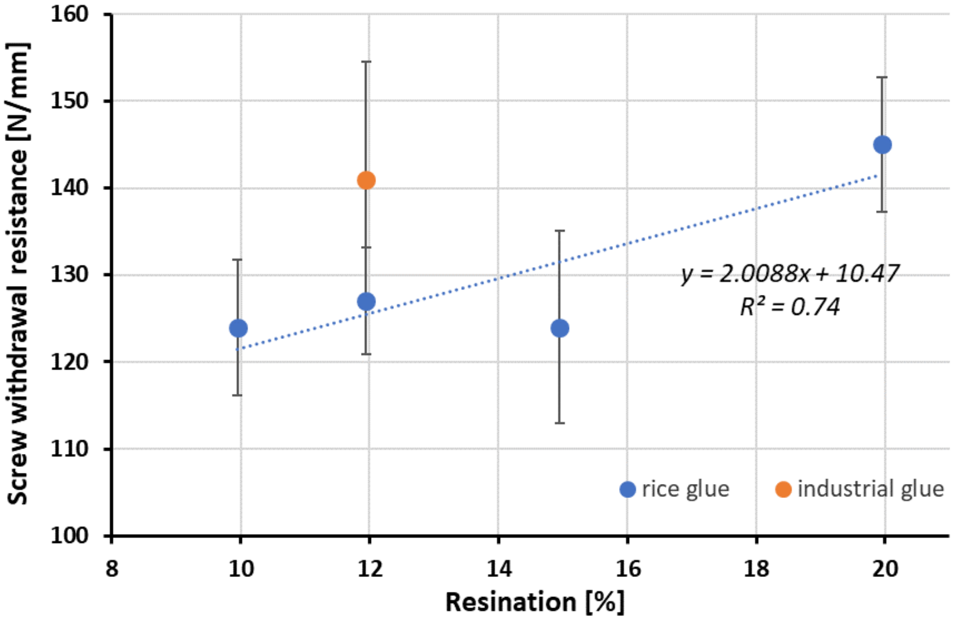

Figure 3. The screw withdrawal resistance of the boards of various content of rice binder

Contact angle

The results of the contact angle test are shown in Figure 4. The chart shows that the contact angle decreases as the time from the drop is placed passes. The results after the 60 s show that the contact angle increases with an increase in the addition of rice starch from R10 to R15 and a significant decrease in the contact angle is shown with the R20 sample. After 60 s, sample R15 shows the highest angle of 93° and has a higher angle obtained on the reference sample of 80°, while R20 shows the lowest angle of 54°. The results for the contact angle after 0 s are very similar for each sample, but the reference sample shows a minimal higher contact angle. Well-chosen proportions of rice starch in the panels can give good hydrophobic properties, presented by the high contact angle. There were no statistically significant differences between the values of the contact angle after 0 s, but after 60 s, all the achieved values varies statistically significantly.

Gumowska and Kowaluk (2023) studied the water drop test where the results for dry starch (DS) and wet starch (WS) samples showed the highest hydrophobicity which could be an effect of the reduced porosity of the HDF boards due to the 20% resination. Thus, as the starch addition increased for 1 s, as well as 60 s, the average contact angle values increased.

Interesting contact angle results came out in the study of Dasiewicz and Kowaluk (2022) where cellulose-based bonded plywood was created. All cellulose samples showed very good hydrophobic properties. The contact angle after 1 s was very similar to that after 60 s in each sample bonded with a biodegradable adhesive. It should be pointed out, that the surface of produced composites was partially thermo-modified, which could contribute to high contact angle.

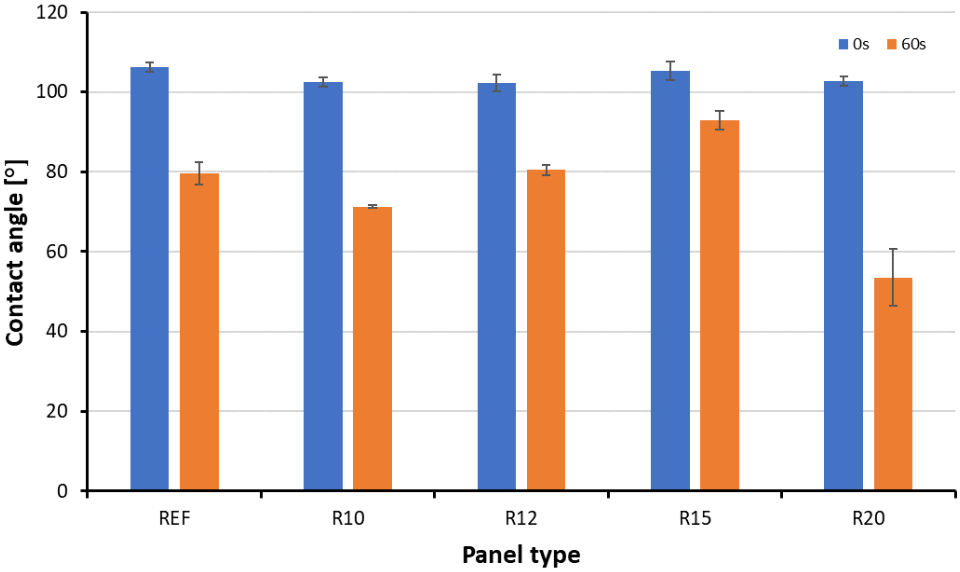

Figure 4. The contact angle of tested samples

Density profile

The results of the density profile are shown in Figure 5. To enhance the readability of the plot, knowing that the density profile is symmetrical along the middle of the thickness, half of the thickness has been presented. For the surface layer, the highest density values were obtained at two different thickness zones. At a thickness of 0.4 mm for sample R15, the density came out to 1006 kg/m^3 at 2.8 mm for sample R20, the density came out to 1004 kg/m^3. In the inner layer, the lowest density came out for the R20 sample at 785 kg/m^3. Such a low density of the core layer caused the low internal bond. It should be also stated, that with the increasing rice starch amount, the face layers' high density zone has moved to the core layers.

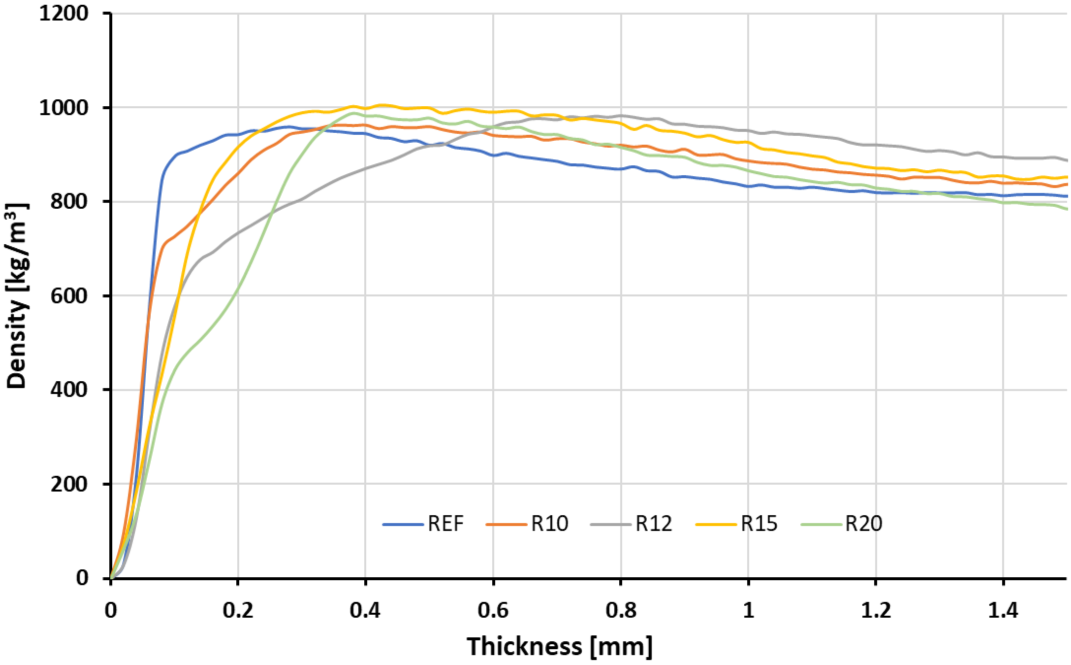

Figure 5. The density profiles of tested samples

Swelling in thickness (TS), Water absorption (WA)

The results of the thickness swelling measurement are shown in Figure 6. Samples bonded with rice glue after 2h and 24h soaking in water show a gradual decrease in swelling with an increase in starch addition. The highest swelling is achieved by the R12 sample after 24h 153% and after 2h 133%, while the lowest swelling is achieved by the R20 sample after 24h 107% and after 2h 90%. However, the reference sample shows the least swelling after 2h and 24h, 32% and 34% respectively. Only the reference samples out of all tested samples meet the requirements of EN 622-5 (2010). The only statistically significant differences between TS average values have been found for 20% resination panels when referring to remaining panels with rice binder.

Feŕndez-García et al. (2012) in their study did not obtain any particleboards with added starch that could meet the standard of TS values for load-bearing capacity (class P4) because the results show that TS values increased with increasing adhesive content. Peterson et al. (2020) as well, since in their study they did not receive good TS and WA results because regardless of the adhesive matrix used, an increase in the concentration of wood flour resulted in an increase in the amount of water absorbed by the composite.

The results of the water absorption measurement are shown in Figure 7. As in TS, absorption decreases with higher rice glue content. As in TS, the absorption decreases with higher rice glue content from sample R10 after 2h 226% and after 24h 252% to sample R20 after 2h 160% and 24h 179%. The lowest water absorption is achieved by the reference sample after 2h and 24h, 80% and 82% respectively. The only statistically significant differences between WA average values have been found for 20% resination panels when referring to remaining panels with rice binder.

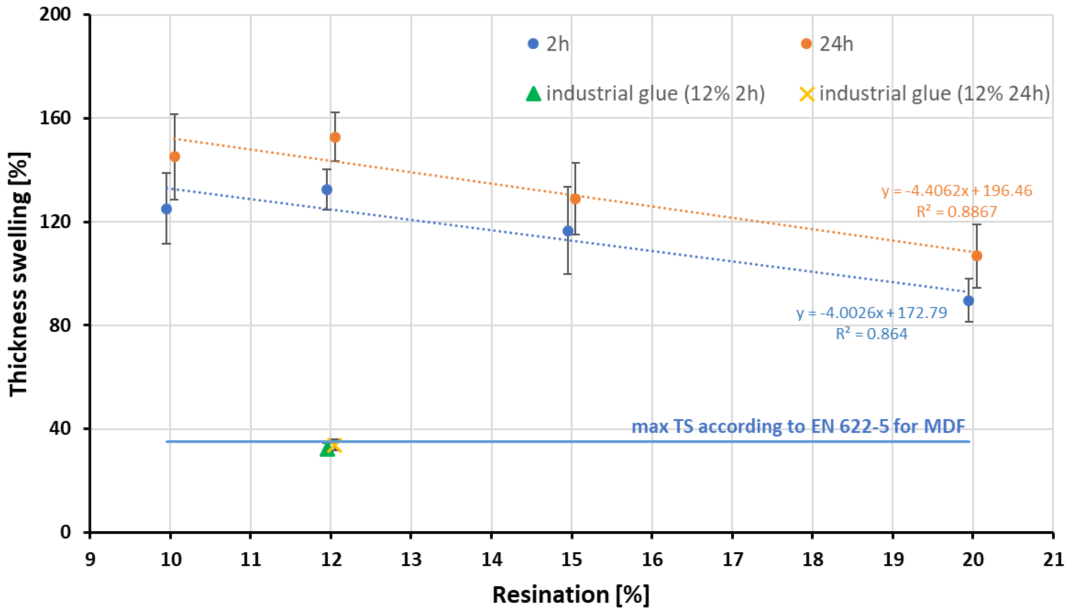

Figure 6. Swelling of the thickness of the boards with different contents of rice glue

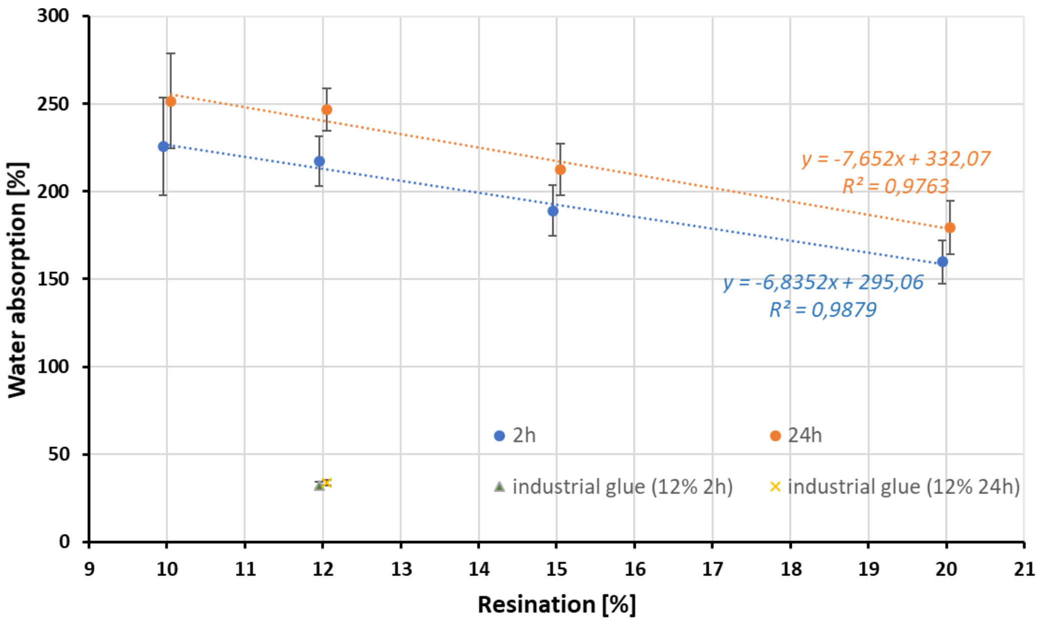

Figure 7. The water absorption of the boards with different contents of rice glue

Surface water absorption (SWA)

The results of surface water absorption are shown in Figure 8. Surface water absorption decreases significantly from 8611 g/m^2 for the R10 sample to 4155 g/m^2 for the R20 sample. However, the lowest SWA was for the reference sample, which obtained 3702 g/m^2. From the results, it can be concluded that the greater the addition of rice starch, the lower the surface water absorption. Gumowska and Kowaluk (2023) in their study obtained decreasing SWA with the increasing biopolymer binder content. However, the lowest average SWA value was recorded for a reference sample glued with UF adhesive.

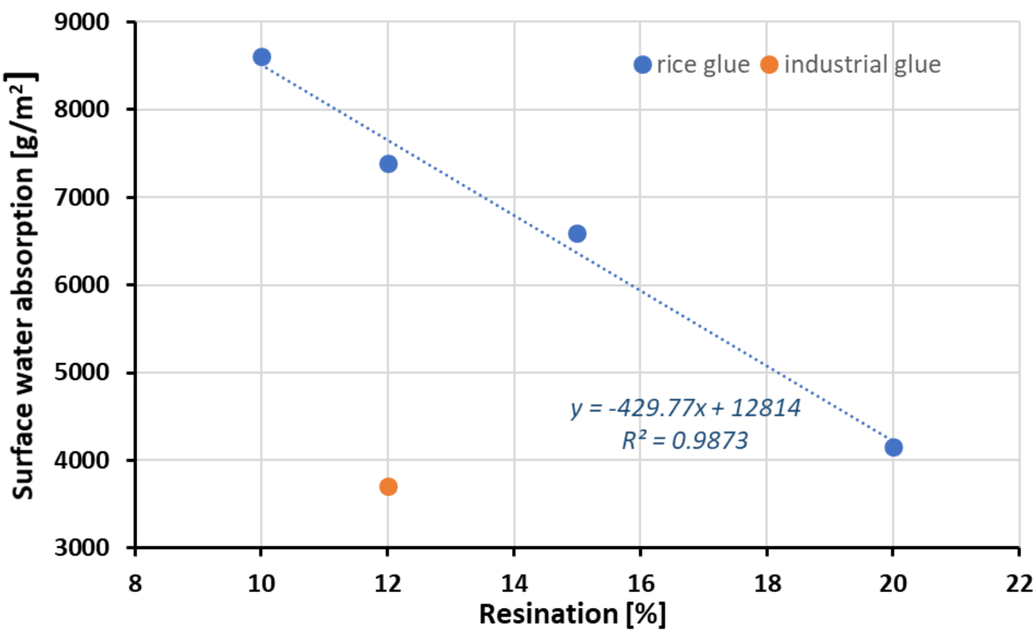

Figure 8. Surface water absorption of tested samples

CONCLUSIONS

According to the conducted research and the analysis of the achieved results, the following conclusions and remarks can be drawn:

1. With the increase in the addition of rice binder, the modulus of rupture and modulus of elasticity gradually increases and reaches values that meet the minimum requirements of the relevant European standards.
2. Internal bonding values with increasing addition of rice binder decreased and did not meet the minimum requirements of European standards.
3. Thickness swelling, water absorption and surface water absorption decreased with increasing rice binder.
4. The contact angle showed very good properties for the R15 sample, which had better properties than the reference sample. So a well-chosen amount of rice starch addition can give good hydrophobic properties.
5. With the increasing rice binder amount, the face layers' high density zone has moved to the core layers.

In conclusion, it is possible to note that rice starch can be recognized as a potential alternative to replace the adhesives previously used in HDF board technology in appropriately selected amounts.

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Streszczenie: *Charakterystyka płyt pilśniowych o wysokiej gęstości produkowanych przy użyciu skrobi ryżowej jako spoiwa*. Kleje stosowane jako spoiwa w płytach nie ulegają biodegradacji, a zawarty w nich formaldehyd jest toksyczny. W dzisiejszych czasach bardzo ważne jest dbanie o środowisko. Istnieje wiele biodegradowalnych produktów, które podobnie jak obecne spoiwa do płyt mogą spełniać dobre właściwości mechaniczne i fizyczne, a jednocześnie nie szkodzić środowisku. W związku z tym w niniejszym projekcie celem badań było określenie możliwości tworzenia płyt pilśniowych formowanych na sucho przy użyciu skrobi ryżowej jako spoiwa. Badania obejmowały produkcję płyt z masowym udziałem skrobi ryżowej 0%, 10%, 12%, 15% i 20% względem masy zupełnie suchych włókien oraz badanie ich właściwości fizycznych i mechanicznych. Wyniki dowiodły, że skrobia ryżowa może być stosowana jako środek wiążący, jeśli wybierzemy odpowiednią ilość skrobi, aby poprawić niektóre właściwości mechaniczne i fizyczne. Bardzo dobre właściwości uzyskano w przypadku określania modułu sprężystości przy zginaniu i wytrzymałości na zginanie oraz odporności na wyciąganie śruby przy wysokiej zawartości spoiwa ryżowego, ale z drugiej strony w przypadku spoin wewnętrznych wysoka zawartość ryżu obniżyła właściwości. W niektórych badaniach mąka ryżowa poprawiła właściwości, ale nie na tyle, aby spełnić wymagania normy, jak miało to miejsce w przypadku spęcznienia na grubość.

Słowa kluczowe: płyta pilśniowa, HDF, skrobia ryżowa, spoiwo

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