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Non-food use of solid residues from the dairy industry as a binder in dry-formed fiberboard technology

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Abstract: *Non-food use of solid residues from the dairy industry as a binder in dry-formed fiberboard technology.* The research investigated the possibility of using solid residues from the dairy industry as a binder in dry-formed fiberboard technology. The scope of work included the production of boards with a mass content of milk powder of 0%, 10%, 12%, 15%, and 20% (concerning the totally dry mass of wood fibers) and studying their selected physical and mechanical properties. The results show that the properties of the produced boards are related to the mass amount of the binder, and that is, using the right amount of binder makes it possible to obtain values that meet the requirements of the relevant European standards.

Keywords: fiberboard, MDF, HDF, binder, milk powder

INTRODUCTION

HDF is a non-load-bearing product for indoor use in dry conditions, as its mechanical strength is low, and it is not resistant to moisture (Badin et al. 2018). This work investigates the possibility of using solid post-production residues from the dairy industry as a binder in dry-formed fiberboard technology. In the scope of the work, test material is planned to be produced under laboratory conditions with different proportions of the mentioned alternative binder. Reference materials will be produced using boards with a commercial binder commonly used in the HDF industry. The produced test material will be subjected to characterization of selected mechanical and physical properties.

The Mary et al. (2024) study investigated the potential of specific raw materials - soybean meal, spent grains from microbreweries, shrimp shells, and skimmed milk powder - as viable protein sources in adhesive development. We can read that the researchers studied replacing petroleum-based ingredients with natural raw materials such as lignins, tannins, and proteins. Of these alternatives, proteins, which are biological macromolecules, are known for their ability to increase adhesion to wood substrates.

Various sources of bio-based adhesives have been developed by Shukla and Cheryan (2001), Beg et al. (2005), Norström et al. (2014). Of great interest is the use of zein, which is derived from glutenous corn meal and is highly effective as a resin/adhesive (Shukla and Cheryan 2001). Zein is an important storage protein (prolamine) present in corn, accounting for 35-40% of its total protein content. It is extracted from gluten meal, obtained by wet milling of corn kernels. Unfortunately, its high cost is an obstacle to most commercial applications, such as the production of bioplastics and biocomposites (Shukla and Cheryan 2001).

Pesenti et al. (2017) used Ulex europaeus in their study to produce non-toxic binder-free fiberboard. Ulex europaeus is one of the most widespread and aggressive invasive plants in the world. Its fibers, which can be obtained through an alkaline dissolution process, have been successfully thermally compressed into high-density fiberboard without the use of a binder. The effects of bioorganic and crystalline components on the final product have been studied using crystallographic, thermoanalytical and mechanical techniques. Despite the presence of residual lignin in small amounts, it contributed to fiber cohesion, improving hydrolysis and adhesion properties. The best overall properties were observed in compressed products with a density of 1030 ± 38 kg/m^3, which showed an elastic modulus of 4.31 ± 0.26 GPa and a modulus of rupture equal to 26.5 ± 1.3 MPa.

Dasiewicz and Kowaluk (2023) conducted a study in which they determined the feasibility of producing dry-formed fiberboard using rice starch as a binder. The results of their research proved that rice starch can be used as a binder if the right amount of starch is chosen to improve certain mechanical and physical properties. Very good properties were obtained for determining the bending modulus and flexural strength, as well as the bolt pullout resistance with a high rice binder content, but on the other hand, for internal bonds, the high rice content lowered the properties.

Currently, a lot of waste is generated, which is very harmful to the environment. Adhesives used as binders in the production of wood-based composites are not biodegradable, and the formaldehyde they contain is toxic. (Dasiewicz and Kowaluk 2023). Therefore, the study conducted aimed to investigate the possibility of using milk powder as a natural binder in the production of panels, which would minimize toxicity from formaldehyde and allow for biodegradation of the panels.

MATERIALS AND METHODS

Materials

The panels were produced under laboratory conditions from industrial softwood pulp of 95% pine (*Pinus sylvestris* L.) and spruce (*Picea abies* (L.) H.Karst) with a moisture content (MC) of about 3%. Milk powder (Okręgowa Spółdzielnia Mleczarska w Siedlcach, Siedlce, Poland) was used as a binder for the panels' production. No other binder was used in the production of the investigated panels.

For reference panels, the industrial urea-formaldehyde (UF) resin with a dry matter content of 65% has been used. The hardening time of the resin mixed with ammonium nitrate hardener at 100°C was 82 seconds, and the resination was 12%.

Production of the panels

HDF fiberboard with a nominal thickness of 3 mm and a nominal density of 800 kg/m^3 was produced under laboratory conditions with two sheets per variant tested.

The application of the dairy agent was successively 10%, 12%, 15%, and 20% concerning the total dry weight of the wood fibers. The pressing parameters were as follows: hydraulic press (AKE, Mariannelund, Sweden), temperature: 200 °C, pressing factor of 20 s/mm of nominal panel thickness, and a maximum unit pressure of 2.5 MPa. The produced panels were conditioned at 20°C ± 1°C and 65% ± 2% relative humidity for 7 days to stabilize the mass before testing.

Characterization of the panels

The following physical properties of the panels were analyzed during the tests: the modulus of elasticity (MOE) and modulus of rupture (MOR) (bending strength) were determined according to EN 310 (1993), the determination of swelling in thickness (TS) and water absorption (WA) after soaking in water according to EN 317 (1993), surface water absorption (SWA) according to EN 382-2 (1993). Tensile strength (IB) according to EN 319 (1993), screw withdrawal resistance (SWR) according to EN 320 (2011), and the water contact angle (CA) were tested on a PHOENIX 300 (SEO Co. Ltd, South Korea) using distilled water, angle measurement was performed 1 s and 60 s after the drop was deposited on the test surface. The density profile was measured using a density profilometer (Laboratory Density Profile Measuring System) from GreCon (Fagus- GreCon Greten GmbH and Co. KG, Alfeld/Hannover, Germany). For all tests of mechanical and physical properties, a minimum of 6 repetitions were performed for each sample type tested. The results presented in the graphs, where used, show the mean values and standard deviations as error bars.

Statistically significant differences between the obtained mean values, where applicable, were distinguished by ANOVA analysis.

RESULTS AND DISCUSSION

Modulus of elasticity

The elastic modulus data for panels with different milk binder contents are shown in Figure 1. Analysis of the results showed that the lowest MOE value occurred for sample M10 and was 2212 N/mm^2. The highest result was for sample M15 2909 N/mm^2, which was higher than the reference sample (REF) 2672 N/mm^2. Interestingly, the sample with the highest dairy binder content (20%) showed a lower elastic modulus than the M15 sample, it was 2474 N/mm^2.

Similar results were obtained by Dasiewicz and Kowaluk (2023) in their study of panels with rice flour binder. As the rice starch content increased, they noted an increase in modulus of elasticity. In the case of their study, also only one sample (M20) recorded a higher MOE value than the reference sample.

In the study by Sulaiman et al. (2013), mechanical properties were recorded for samples bonded with modified starch. The highest average MOE value for panels with higher density (0.80 g/cm^3) was 3471.64 N/mm^2. The only M15 samples met the minimum requirements of EN 622-5 (2009), which is 2700 N/mm^2. However, it should be pointed out that such a level is given for panels of the thickness in the range of 4-6 mm (no minimum MOE requirements for panels below 4 mm).

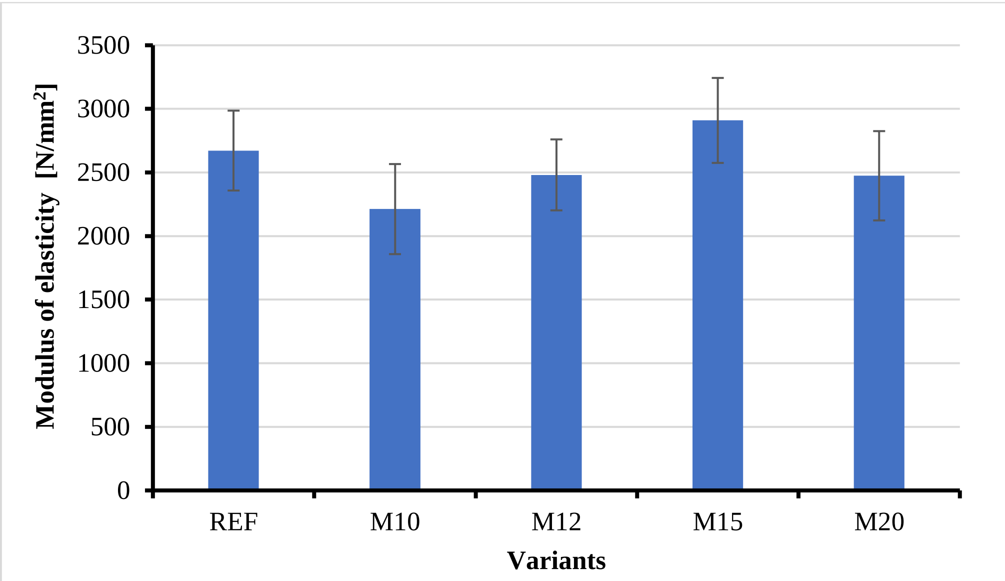

Figure 1. Modulus of elasticity of panels with different milk binder content

Modulus of rupture

The results of measuring the bending strength of fiberboard with different milk binder contents are shown in Figure 2. The results, as with MOE, show that the lowest value was recorded for the M10 variant which was 24.0 N/mm^2. The highest MOR result was also recorded for sample M15 35.0 N/mm^2. Interestingly, the reference sample (REF) recorded the same value of 35.0 N/mm^2.

Similar results were obtained by Wronka et al. (2020), where in their study, they created the panels bonded with potato starch. Hazim et al. (2020) also obtained similar values when testing the panels sealed with citric acid-modified corn starch. In these studies, the MOR value increased with increasing binder addition. The MOR results show that all the tested panels reached values that met the minimum requirements of EN 622-5 (2009).

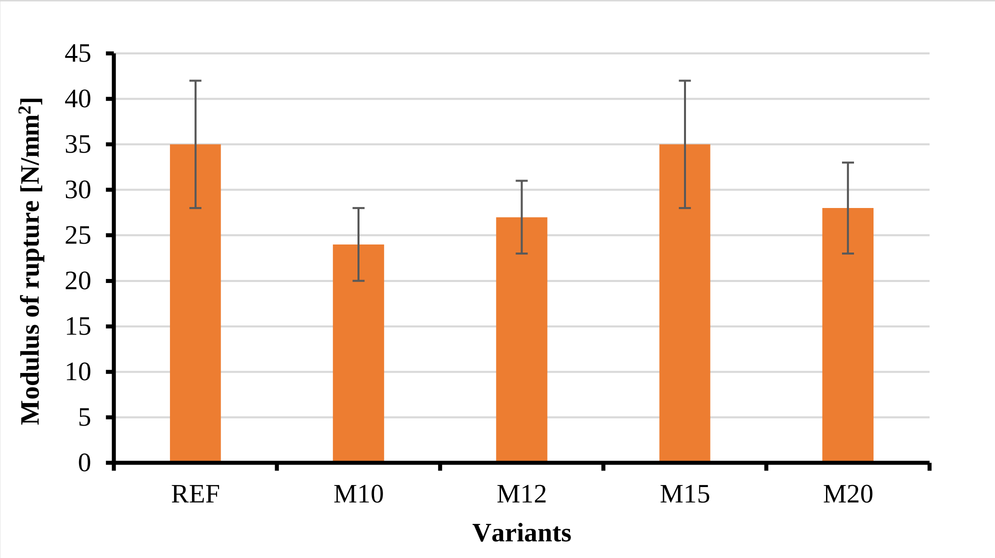

Figure 2. Modulus of rupture of panels with different milk binder content

Water absorption

The results of water absorption of fiberboard with different dairy binder contents are shown in Figure 3. The values are shown from measurements taken after 2h and after 24h of soaking the samples in water.

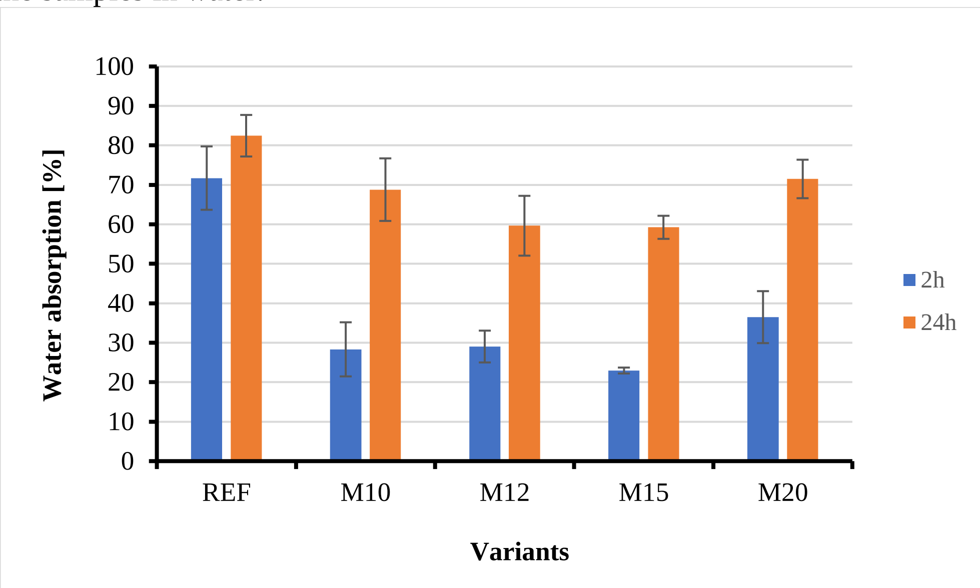

Figure 3. The water absorption of the boards with different milk binder contents

From the graphical data, we can read that water absorption recorded much lower values for the samples sealed with dairy binder than for the reference sample (REF) 31.4% after 2h and 82.5% after 24h. The smallest WA value was recorded for variant M15, with a value of 23% after 2h and 59.2% after 24h. The largest, however, interestingly, was the M20 sample, whose value was 36.5% after 2h and 71.5% after 24h.

It is worth noting that even with the smallest variant M10, WA results were much lower than for the REF sample, with 28.3% after 2h and 68.8% after 24h.

Interesting WA results were obtained by Borysiewicz and Kowaluk (2023), who studied selected properties of MDF boards glued with different fractions of recycled HDPE particles. The researchers obtained the lowest water absorption results for fractions below 1 mm. They assumed that the reason for such results may have been due to the homogeneity of these panels since as the HDPE binder size increased, there seemed to be more spaces filled exclusively with wood fibers, which can absorb water more intensively.

Thickness swelling

The results of swelling per thickness of fiberboard with different milk binder contents are shown in Figure 4. Analysis of the results shows that as the milk binder content increases, the swelling of the board decreases. After both 2-hour and 24-hour soaking, the lowest swelling was recorded for the sample with 15% binder content. After 2 h, it was 12.8%, while after 24 h, it was 27.1%. It is worth noting that even with the lowest content of milk powder (10%), the samples after the 2-hour soak recorded lower values than the reference sample (REF). The samples after 2h showed a result of 19.5% while the reference sample showed 28.3%. However, after 24h, the values changed, as both sample M10 (34.7%) and sample M12 (33.6%) recorded swelling higher than the reference sample (31.4%).

It is noteworthy that after 24 hours of soaking, sample M20 records greater swelling than the reference sample, and after both phases of soaking greater than each of the previous samples tested, the percentage of this binder was lower. The M20 sample records 21.2% after a 2-hour soaking and 36.1% after 24 hours. These results may suggest to us that the tested binder exhibits the desired values only with the application of appropriate sealing ratios.

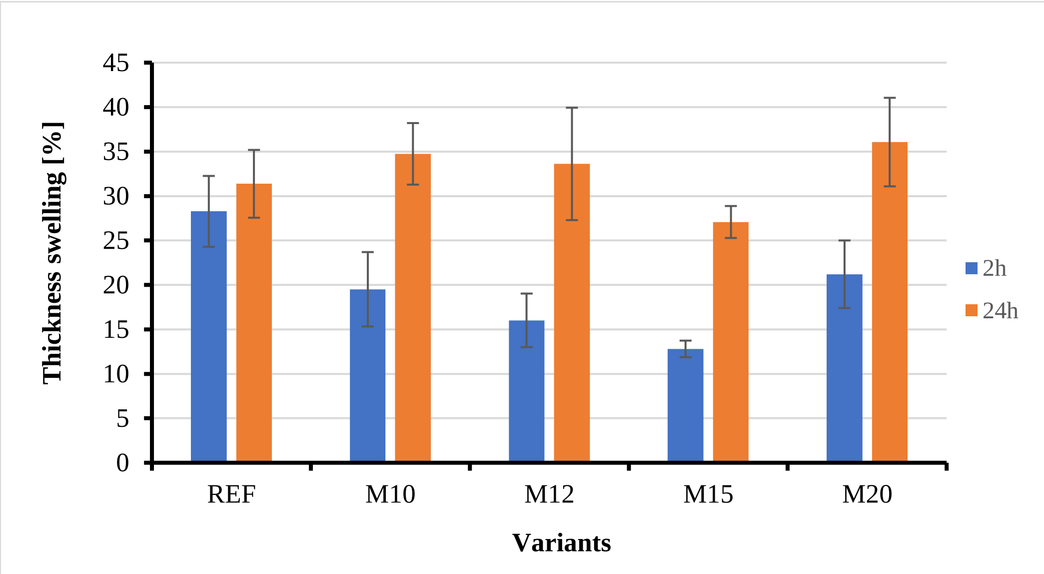

Figure 4. Thickness swelling by the boards with different milk binder contents

All the results obtained reach values that meet the requirements of EN 622-5 (2009). Similar results were obtained in a study by Bartoszuk and Wronka (2023), which investigated the effect of recycled artificial leather particle content in particleboard. This research showed that as the leather content of the boards tested increased, swelling decreased.

Interesting results were obtained in a study by Pawlik and Kowaluk (2023). The effect of two types of release agents on selected properties of fiberboard was studied there. The results showed that swelling decreased with an increase in the amount of the applied formulation for both agents “A” and “B”. There, too, with agent “A”, the samples with the smallest application (10 g/m^2) showed a swelling lower than that of the reference sample (REF) and is 31.3% after 2h and 32.1% after 24h.

Surface water absorption

The results of the surface absorption of panels with different milk binder contents are shown in Figure 5. As can be seen, SWA gradually decreases as the amount of binder increases. The highest value of surface absorption was recorded for sample M10, which was 752 g/m^2. The smallest recorded value was 171 g/m^2 for sample M20. It is worth noting that the difference in SWA between the reference sample (REF) and the M10 sample is very large, the SWA for the reference sample was 3122 g/m^2.

Gumowska and Kowaluk (2023) in their study also obtained decreasing SWA with increasing biopolymer binder content. However, in the case of their study, the lowest surface absorption value was recorded for the reference sample, which was sealed with UF adhesive.

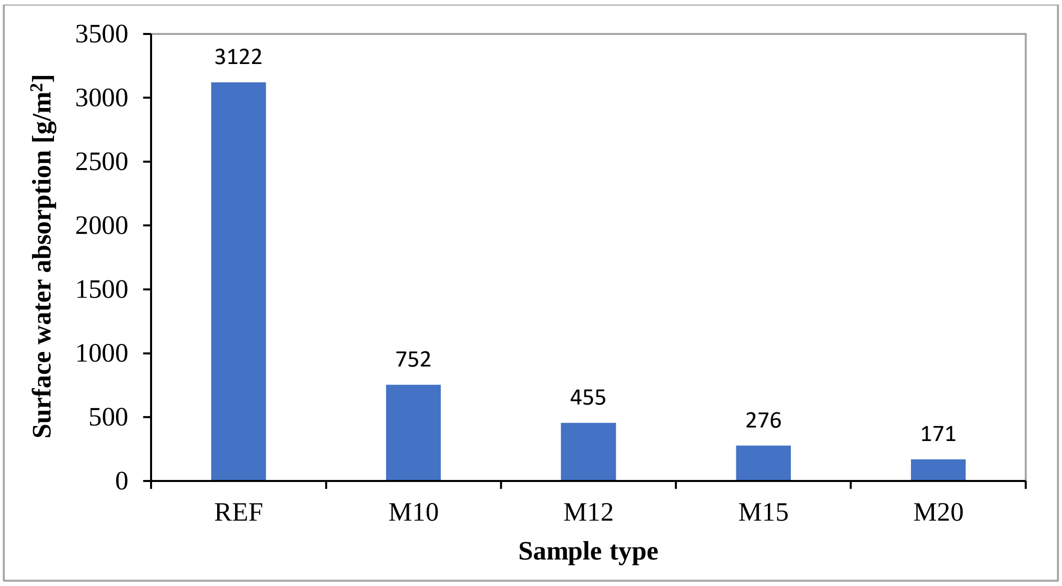

Figure 5. Surface water absorption by the boards with different milk binder contents

Contact angle

The results of the wetting angle test are shown in Figure 6. The graph shows that the wetting angle decreases with time after drop placement. The results after 60 s show that the wetting angle increases as the addition of milk powder increases from M10. After 60 s, the M20 sample shows the highest angle of 87° and has a higher angle than that obtained in the reference sample (REF) of 75°. The lowest angle after 60 s is shown by the M10 sample, whose value equals 68°.

The results for the wetting angle after 0 s are similar for each of the samples tested, but it is the reference sample that shows a minimally higher wetting angle. The study suggests that properly selected proportions of milk powder in the panels can provide good hydrophobic properties.

No statistically significant differences were observed in the wetting angle values after 0 s; however, after 60 s, the values obtained are statistically significantly different.

Gumowska and Kowaluk (2023) conducted a water droplet test in which dry starch (DS) and wet starch (WS) samples achieved the highest level of hydrophobicity. This result can be attributed to a reduction in the porosity of the HDF boards due to the presence of 20% resin. Accordingly, an increase in the average wetting angle values was observed with an increase in the amount of added starch at 1 s and 60 s periods.

Interesting results regarding wetting angle were obtained in a study by Dasiewicz and Kowaluk (2022), which created plywood glued with cellulose glue. All cellulose samples tested showed excellent hydrophobic properties. The wetting angle after 1 s was almost identical to that after 60 s for each sample bonded with biodegradable glue.

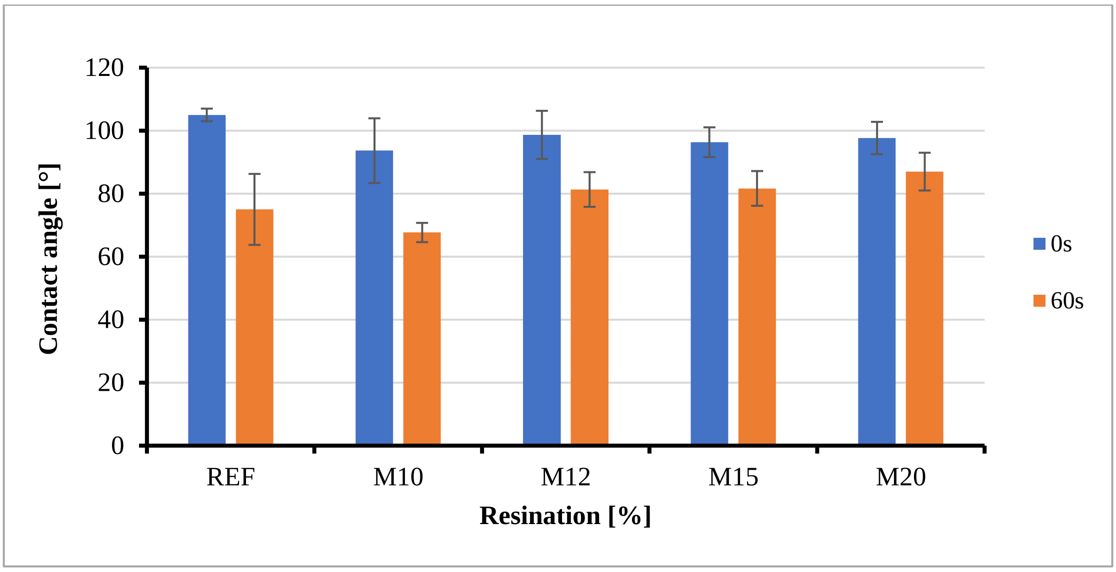

Figure 6. The contact angle by the boards with different milk binder contents

Density profile

The results of the density profile are shown in Figure 7. Analyzing the results of the density of the panels, we can read that the highest density values were recorded for sample M15 where, at a thickness of 2.98 mm of the panel, the density was 1108 kg/m^3, while the lowest value was recorded for sample M12, it was 36.5 kg/m^3 at a thickness of 0.02 mm.

The graph revealed a significant asymmetry, which is probably due to the milk powder spilling over to one side during panel forming, especially at larger seals.

An interesting study was conducted by Borysiuk et al. (2019), who investigated the effect of filler on the density profile of polymer-wood composites. Based on this research, composites with 40-60% lignocellulosic filler content showed a uniform flat density profile characterized by WPC composite panels with 40% filler content. In contrast, an increase in lignocellulosic filler content (50-60%) resulted in a decrease in density in the middle zone of the panel.

The results of the internal bonding of the manufactured panels glued with milk powder are shown in Figure 8. The lowest IB result obtained for sample M10 was 0.40 N/mm^2. In contrast, the highest result was shown by sample M15 - 1.14 N/mm^2. Again, we see in the graph that the sample with the highest milk powder content records lower properties than the other samples.

Once again, this suggests to us that using such a binder in the right proportions will show good mechanical properties.

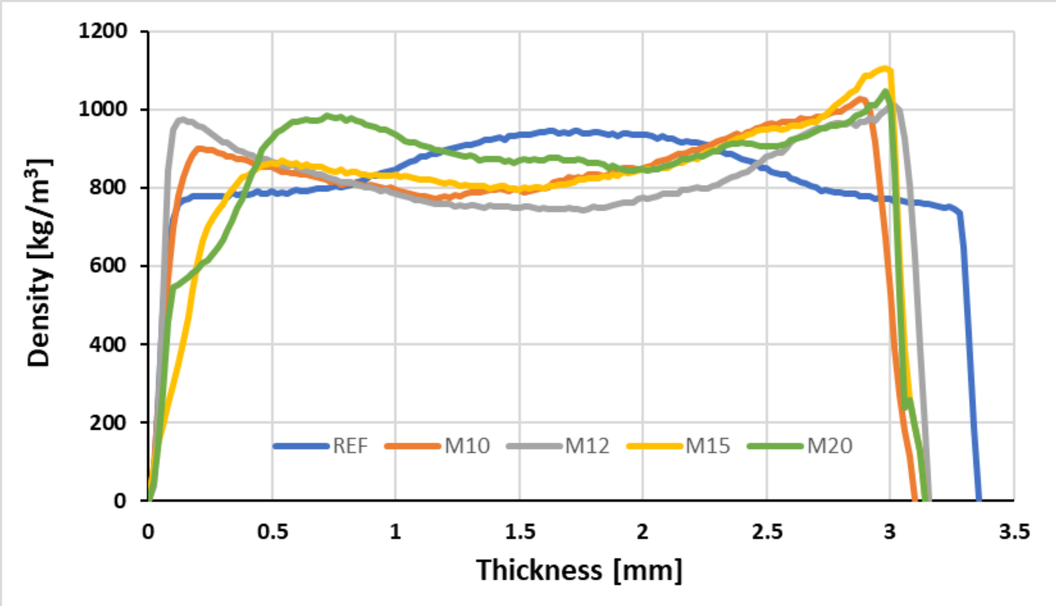

Figure 7. The density profiles by the boards with different milk binder contents

Internal bond

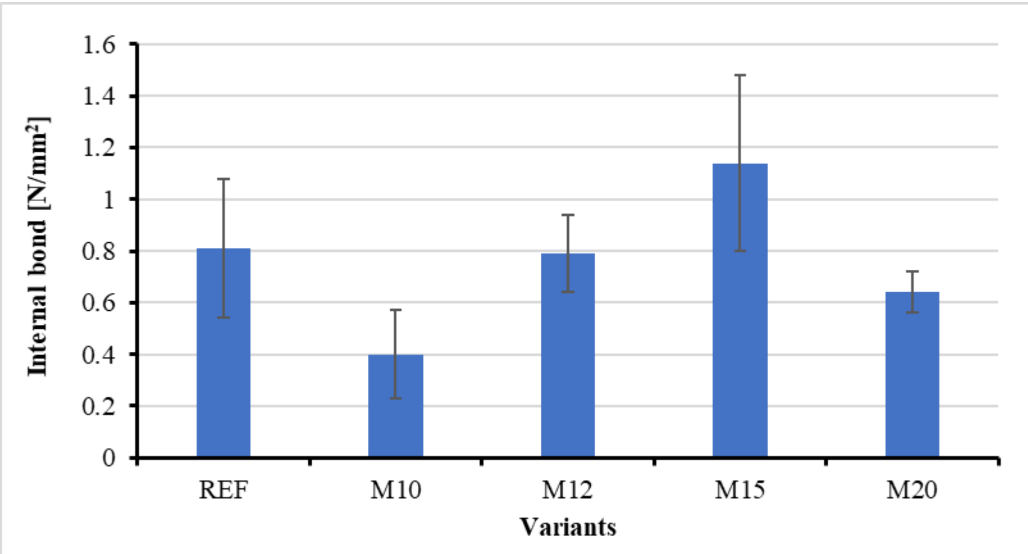

Figure 8. The internal bond of the boards with different milk binder contents

The graph shows that REF, M12, and M15 samples glued with dairy binder have higher values than EN 622-5 (2010) requirements for MDF panels (0.65 N/mm^2). Similar results were obtained during a study on citric acid-modified starch in wood composite production (Hazim et al. 2020), where all tested wood composite samples met the IB requirements according to the specified standard. In contrast, a study by Theng et al. (2017) showed lower IB properties for lignin-added fiberboard.

Screw withdrawal resistance

The results of screw withdrawal resistance from panels with different contents of milk powder are shown in Figure 9. The lowest SWR results were obtained for sample M10, whose value was 90 N/mm, and the highest results were obtained for sample M20 – 105 N/mm. The values obtained are, unfortunately, much lower compared to the values that the reference plate (REF), which had a value of 166 N/mm, obtained in the test.

In the results of Rosa and Kowaluk (2022), we can read that increasing the amount of resin leads to a significant increase in SWR when testing medium-density fiberboard glued with vegetable glue. Nevertheless, also for their study, no sample with plant glue reached higher values than the reference sample.

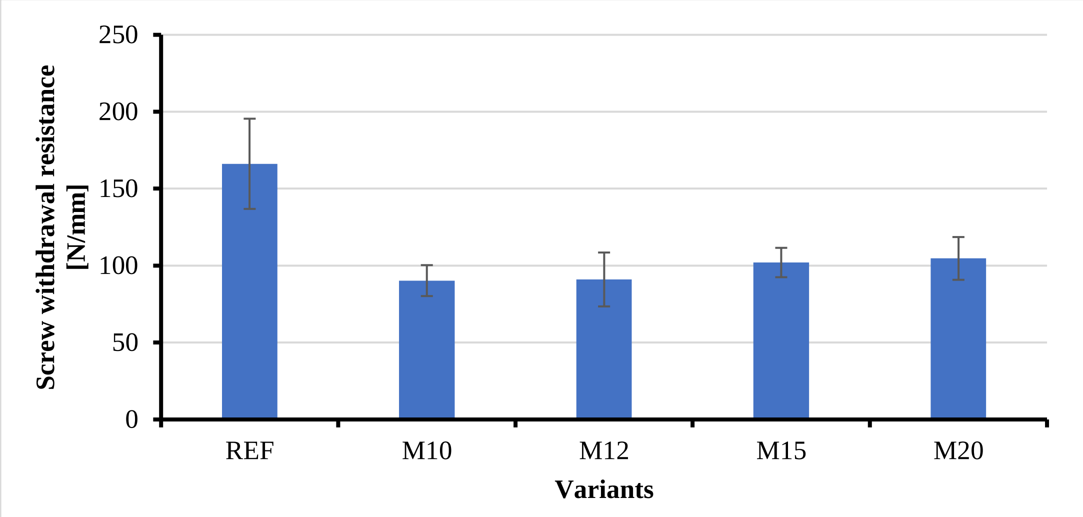

Figure 9. The screw withdrawal resistance of the boards with different milk binder contents

Tab. 1. The statistical assessment results of mean values

Test type	Alternative binder content [%]				
	REF	10	12	15	20
MOE	a*	a	a	a	a
MOR	a	b	a, b	a	a, b
IB	a	b	a	a	a
SWR	a	b	b	b	b
TS 2h	a	b	b	c	b
TS 24h	a	a	a	a, b	a
WA 2h	a	b	b	b, c	b
WA 24h	a	a, b	b	b, c	b
CA 0s	a	a, b	a	b	b
CA 60s	a	a, b	a	a	a

* *a, b… homogeneous group*

CONCLUSIONS

Based on the tests conducted and the analysis of the results obtained, the following conclusions and observations can be drawn:

- As the amount of binder in the form of milk powder is increased from 10%, the modulus of rupture and modulus of elasticity gradually increase until 15% milk powder binder content, where the MOR and MOE reach maximum values. Further increase of alternative binder causes a decrease of mentioned features.
- Swelling thickness and water absorption decrease with alternative binder content increase to 15%, which is especially visible for 2h soaking. The increase in soaking time and the increase of alternative binder content leads to more similar parameters for all tested panels.

- The water contact angle was highest for the M20 sample, which showed better properties than the reference sample. This means that a well-chosen amount of milk powder additive can provide better hydrophobic properties.
- Panel screw withdrawal resistance tests showed that samples with milk powder binder showed poorer mechanical properties than the reference sample.
- The best internal bond strength, which has been higher than for reference panels, has been reached for 15% alternative binder content.
- Increasing alternative binder content from 10% to 20% significantly reduces the surface water absorption.

In conclusion, milk powder can be considered an alternative to replace the adhesives previously used in HDF board technology. If appropriate amounts of milk powder are used, it is possible to obtain panels with the desired mechanical and physical properties.

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Streszczenie: *Nieżywnościowe wykorzystanie stałych pozostałości z przemysłu mleczarskiego jako spoiwa w technologii płyt pilśniowych suchoformowanych*. Celem badań było określenie możliwości wykorzystania stałych pozostałości poprodukcyjnych z przemysłu mleczarskiego jako spoiwa w technologii płyt pilśniowych suchoformowanych. Zakres prac obejmował wytworzenie płyt o udziale masowym mleka w proszku 0%, 10% 12%, 15% i 20% (w odniesieniu do masy całkowicie suchych włókien drzewnych) oraz zbadanie ich wybranych właściwości fizycznych i mechanicznych. Uzyskane wyniki pokazują, że właściwości wytworzonych płyt są związane z ilością masową spoiwa i że stosując odpowiednią ich ilość można uzyskać wartości spełniające wymagania odpowiednich norm europejskich.

Słowa kluczowe: płyta pilśniowa, MDF, HDF, spoiwo, mleko w proszku

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