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A Study of the Properties of UV-Aged and Low Formaldehyde Emissions Particleboards Manufactured with Bio-Based Wood Protein Adhesives

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Abstract: The environmental crisis and the safeguarding of the population's health has led to research into different ways of mitigating harmful gases. Among the emissions that the wood industry has sought to reduce are those of formaldehyde, which is why new green adhesive methods for wood panels have been investigated in recent years. In this research, particleboard with two bio-based wood adhesive (PB-bbwa) formulations. The first PB-bbwa formulation, based on proteins obtained from compounds from the alcoholic beverage industry, and the second PB-bbwa formulation, based on proteins from a mixture of compounds from the alcoholic beverage and food industries, were manufactured and tested to evaluate the physical–mechanical, thermal and formaldehyde emission properties of untreated and UV-treated formulations at a laboratory scale. The results of the physical properties obtained in the PB-bbwa were similar or even better than those of the control PB. Additionally, PB-bbwas improve on the control PB sample's Janka hardness by least 28%, and a decrease in thermal conductivity in the edgewise position and formaldehyde emissions by 12% and 88%, respectively, in comparison to the control PB. The tests performed evidenced that PB-bbwas showed comparable performance against the control PB made with urea-formaldehyde and satisfied international standard requirements.

Keywords: bio-based wood adhesives; formaldehyde emissions; particleboards; urea-formaldehyde

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1. Introduction

The first patents for the manufacture of particleboards (PBs) were registered at the end of the 19th century, but it was not until 1941 that the first factories producing furniture boards were set up in Germany and Switzerland. Immediately after World War II, the production of this type of board increased significantly and spread to several countries [1]. Today PBs are a very popular and common material for the manufacture of all kinds of interior furniture. It can be safely stated that, together with medium-density fiberboards (MDF), they have replaced solid wood as the main material for making furniture [2].

Particleboard is manufactured by subjecting wood particles to pressure and heat, usually sprayed with a synthetic resin. The particles that can be used are residues from other production processes such as planning chips, veneer offcuts, and sawmill chips, as well as products prepared with special machinery such as filaments, flakes or wood strips [1].

Hot pressing, where the mixture of wood particles and resin is compacted to the desired density and thickness, allows the resin to polycondensation to bind the particles or fibers and stabilize the board. Particleboards can have 3 to 5 layers with particles of

different thicknesses arranged so that the layers with the thickest particles are in the center and those with the finest particles are on the faces. This improves the mechanical strength and appearance of the boards [1].

Urea-formaldehyde (UF) resins are widely used in PB manufacturing. This composite is a source of formaldehyde, whose emissions are mainly released from the resin when it is heated in the pressing zone and, to a lesser extent, in the cooling zone of the board. It is in these stages of the process where there is a greater probability of exposure [3].

Petroleum-derived adhesives have long been utilized in particleboard production. Nevertheless, their use presents two major drawbacks. The first is related to the health of users, since the use of formaldehyde-based adhesives has generated controversy due to the emission of carcinogenic compounds in their manufacturing process and after their production. The second problem is associated with production costs, due to the increase in the value of the raw material, in this case petroleum where, in the last 10 years, the rise in prices is due, among other reasons, to the growing demand for this resource for its use as a raw material in the development of various products and its energy application [4].

Currently, despite the existence of international regulations and standards that regulate formaldehyde emissions in wood-based composites products; the scientific community is still concerned about further reducing these emissions levels, carrying out studies that incorporate new technologies to produce environmentally friendly products.

In recent years, the development of bio-based adhesives has increased considerably, so that different materials such as lignin, starch, vegetable proteins, tannins, vegetable oils, and mollusk and barnacle proteins, among others, have been explored with the aim of obtaining substances capable of matching the mechanical properties, thermal stability, bonding performance (strength and types of chemical bonds between the adhesive and the surface), and water resistance offered by traditional adhesives [5].

In 2012, Gu et al. [6] investigated a new formaldehyde-free wood adhesive based on soybean flour and a curing agent (CA). Ammonium hydroxide and epichlorohydrin were added for the preparation of the curing agent. With the newly manufactured adhesive, particle boards were manufactured and evaluated. During the same year, Kadimaliev et al. [7] developed environmentally friendly adhesive formulations from brewing waste. Chemical treatment of the yeast waste increased their adhesive characteristics and their chemical crosslinking, as well as their biological crosslinking, and improved the moisture resistance of the adhesive formulations. In 2013, Londoño et al. [8] fabricated PB samples bonded with different formulations of soy-protein-based adhesives that were tested to evaluate their physical–mechanical properties. Muttila et al., in 2014 [9], explored bio-based eco-friendly adhesives for plywood manufacturing, using soybean meal, phenol, white gypsum, and an agricultural-based powder as binder, which was compared to conventional phenol formaldehyde resin-based plywood. In 2015, Lagel et al. [10] incorporated hydrolyzed wheat gluten in a phenol-formaldehyde synthesis, up to 30%. In addition, the presence of triacetin in the phenol-trigo-formaldehyde formulation improved the bending properties. The hydrolyzed gluten was also tested with glyoxal, as well as formaldehyde, and then mixed with tannin/hexamine resin or pMDI to form a renewable component adhesive. During the same year, Kadimaliev et al. [11] made reconstituted pressed wood from sawdust waste, using modified yeast biomass waste as a biological adhesive, and ultra-dispersed bacterial cellulose (UBC) as a binder and crosslinker.

On the other hand, Cheng et al., in 2016 [12], studied the influence of some additives on the performance of soybean and cottonseed protein-based adhesives, also compared the bond strength of these two types of protein-based adhesives on different wood veneers with a wide range of additives. The same author also investigated the impact of various polysaccharide fillers on the performance of soybean and cottonseed protein adhesive [13]. Núñez et al., in 2016 [14], investigated the use of yeast single-cell protein extract (SCPey) as a raw material to produce innovative wood adhesives. Wood veneers glued with the adhesive solutions were used for mechanical testing and adhesive performance evaluation. In 2017, Liu et al. [15] applied different pH conditions to catalyze the reaction of condensed

tannins (CT) and soy protein isolate (SPI) to develop a thermosetting resin. The resulting resin was applied in the manufacture of plywood and its wet shear strength was measured. In 2018, Núñez et al. [16] formulated adhesives based on Rhodotorula rubra (Rr) yeast protein extract to make particleboard (PB). The results were compared with a commercial urea-formaldehyde (UF) adhesive. The results of this research validated the manufacture of particleboard with adhesives based on Rhodotorula rubra yeast protein extract, where the PBs obtained showed similar characteristics to traditional PBs (made with UF), with the advantage that those made with adhesives based on Rhodotorula rubra yeast protein extract met the high formaldehyde emission standards required by markets that aim at sustainable products. During the same year, Ghahri and Pizzi [17] tried to improve their soy-protein-based adhesives with the addition of tannins. In 2019, Bai et al. [18] developed a yeast hydrolysate-based wood adhesive by adding sodium dodecyl sulfate (SDS), polyvinyl alcohol, and ethylene glycol diglycidyl ether (EGDE), to improve its properties through the epoxy group EGDE. Núñez et al., during the same year [19], conducted a preliminary investigation of wood adhesives based on Rhodotorula rubra yeast protein extract reinforced with resorcinol and glyoxal. In 2021, Fagbemi and Sithole [20] conducted a study investigating the potential benefit of keratin protein hydrolysate extracted from chicken feather waste biomass as a bio-adhesive for particleboard production.

The aim of this study is to address the problems associated with urea-formaldehyde-based adhesives, which present a risk to human health because they are the main source of formaldehyde emissions in particleboards. In addition, wood particleboards have limited mechanical performance, which limits their application in structural fields.

Given the above, the main objective of this research is to explore the potential of PBs manufactured with bio-based wood adhesives, with reduced formaldehyde emissions as a solution to safeguard human health. In addition, it is intended to evaluate the feasibility of using these adhesives to maintain or improve the mechanical performance of commercially available panels.

2. Materials and Methods

2.1. Materials

Three-layer particleboard (PB) was manufactured, with fine-sized *Pinus radiata* D. DON wood particles (thickness from 0.4 to 2 mm) on both surfaces and a layer of thick particles (thickness from 2 to 10 mm) in the center, which had an average moisture content of 8%. The PBs were made with urea-formaldehyde (UF) adhesive and with two bio-adhesives based on yeast protein, lignin and MDI. The dimensions of the panels were 500 mm long × 500 mm wide and 12 mm thick. The wood particles, the UF adhesive, and the two bio-based wood adhesives were provided by the Laboratory of Engineering Products and Adhesives based on Wood (PRODIMA-LAB), of the Universidad del Bío-Bío, Concepción, Chile.

The PBs with UF adhesive were manufactured with a UF-S adhesive for fine particles (surface layers) and with a UF-M adhesive for coarse particles (middle layer). The bio-based wood adhesives were manufactured with yeast proteins, lignin, and MDI in a ratio of 70/20/5 (*w*/*w*/*w*) for the first bio-based wood adhesive (P1), and in a ratio of 90/5/5 for the second bio-based wood adhesive (P2), according to the preparation method and mixing temperature [16]. The yeast proteins, lignin, and MDI were provided by the PRODIMA-LAB, Concepción, Chile.

The utilized adhesives properties are listed in Table 1.

Table 1. Properties of the adhesives in the data sheets.

ID	Viscosity (cP)	pH	Electrical Conductivity (mS)	Solid Content (%)
UF-S	387	8.2	0.37	67.51
UF-M	487	8.3	4.27	67.35
P1	406	5.5	3.59	51.87
P2	198	5.7	12.05	49.11

2.2. Manufacture of Particleboards Using Commercially Available Adhesive and Bio-Based Wood Adhesives

Five replicates of each of the three different types of PBs were manufactured. The summary of the samples is listed in Table 2.

Table 2. Summary of the particleboard samples manufactured in this research.

ID	Replicates	Particleboard Dimensions	Amount of Adhesive (g)
C	5	40 cm × 40 cm × 1 cm	101.0
P1	5	40 cm × 40 cm × 1 cm	131.2
P2	5	40 cm × 40 cm × 1 cm	138.6

The pressing parameters were the same in the 3 samples. The gluing ratio was 10%, the pressing factor was 0.8 (mm/min), the particle composition of each PB was 60% for the middle thick layer, 20% for one surface thin layer, and 20% for the other one surface thin layer.

The amount of adhesive used in the manufacture of each board was determined on the basis of an initial CH of the particles of between 2 and 4%, to obtain boards with a final CH of between 8 and 10%.

After conforming the panels, they were pre-pressed and hot pressed. The 2–3 bar pre-pressing was carried out at room temperature at 20 ± 3 °C. After that, a hot pressing was carried out at a temperature of 180 ± 3 °C, by means of a three-stage pressing cycle shown in Figure 1 for the control sample and in Figure 2 for the samples made of bio-based wood adhesives. The difference between them was the pressing time used. The PB based on bio wood adhesive needed more pressing time to evaporate their high-water quantity than the PB based on UF adhesive.

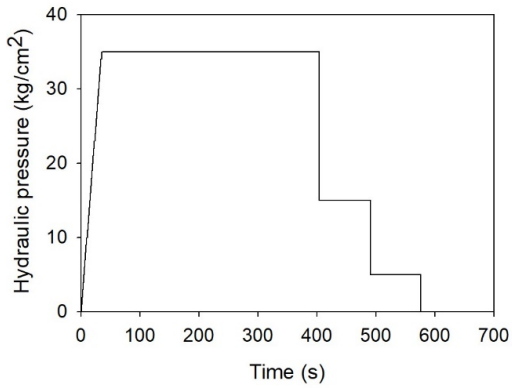

Figure 1. Hot three-stage pressing cycle for control PB samples.

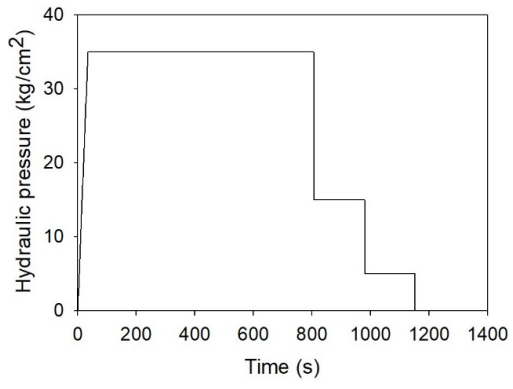

Figure 2. Hot three-stage pressing cycle for samples based on bio wood adhesives.

Immediately after pressing, PBs were kept at room temperature conditions for 7 days. Finally, PBs were cut to determine their physical, mechanical, thermal properties and formaldehyde emissions according to European and American standards' specifications.

2.3. Evaluation of Accelerated UV Aging

The evaluation of accelerated UV aging was performed on 3 specimens of each sample with dimensions of 300 mm length × 50 mm width to be exposed to a continuous aging cycle of UV, according to the American standard ASTM G154 [21].

To analyze the influence of color change (photo discoloration) and the surface appearance of the tested boards on the different adhesives used, a "general factorial" experimental design was carried out using Design Expert® V10.01 software. The factors analyzed were the adhesive and the type of board, and the response variable was the CIELAB color change (Figure 3).

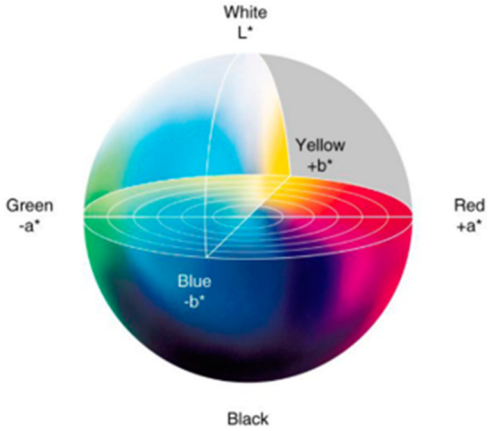

Figure 3. Model for objective color measurement of the International Commission on Illumination (CIELAB). Its color coordinates, L*a*b*, describe lightness: brightness (L*+) and black (L*−), and the colors: red (a*+), green (a*−), yellow (b*+), and blue (b*−) [22].

This test allowed a comparison between the results of the physical and mechanical properties of the normal (untreated) samples and the accelerated UV aging subjected samples (treated samples), to evaluate possible photodegradation.

2.4. Physical Properties

In first place, density was measured in 6 specimens of each sample with dimensions of 50 mm length × 50 mm width, according to the European standard UNE-EN 323 [23]. The density profile of the panels specimens was measured by using an Amersham plc AMCK6693 gamma-ray densimeter, scanning directly through the thickness of the sample with an incremental step of 0.1 mm. A density profile of 2 specimens of each sample with dimensions of 50 mm length × 50 mm width of each PB samples was recorded.

Moisture content was measured in 4 specimens of each sample in accordance with the European standard UNE-EN 322 [24] and, finally, thickness swelling and water absorption were determined in 8 specimens of each sample with dimensions of 50 mm length × 50 mm width under the conditions of the European standard EN 317 [25] for 24 and 48 h.

Physical properties were measured on untreated samples and UV-treated samples. Then, they were compared to evaluate possible photodegradation on the treated samples.

2.5. Mechanical Properties

2.5.1. Static Bending Test

The bending properties of the PB samples were determined though mechanical tests of three-point static bending, according to the specifications and methodology of the European standard UNE-EN 310 [26]. A total of 12 specimens were tested for each sample,

with dimensions of 290 mm length × 50 mm width. The type of failure produced in the specimens was observed and classified using the European standard EN 310 [26].

Stiffness and flexural strength properties were measured on untreated samples and UV-treated samples. Then, they were compared to evaluate possible photodegradation on the treated samples.

2.5.2. Tensile Strength

The tensile strength test of PB was performed on 7 specimens of each sample with dimensions according to the American standard ASTM D3500 method A [27].

2.5.3. Internal Bond Strength

An internal bond (IB) strength test was performed on 8 specimens of each sample with dimensions of 50 mm length × 50 mm width × 12 mm thick, according to the European standard EN 319 [28].

The internal bond strength property was measured on untreated samples and samples subjected to accelerated UV aging. Then, they were compared to evaluate possible photodegradation on the treated samples.

2.5.4. Hardness Test

The Janka hardness test of PB was performed on 6 specimens with dimensions of 100 mm length × 50 mm width, according to the American standard ASTM D1037-12 [29].

Mechanical properties were measured using a universal testing machine, Instron EMIC model 23–100, equipped with BlueHill2 software.

The Janka hardness property was measured on untreated samples and samples subjected to accelerated UV aging. Then, they were compared to evaluate possible photodegradation on the treated samples.

2.6. Thermal Properties

A thermal conductivity test of PB was performed on 6 specimens of each sample, with dimensions of 100 mm length × 50 mm width in flatwise and edgewise positions, using a Decagon KD2 PRO, according to the American standard ASTM D5334 [30].

2.7. Evaluation of Formaldehyde Emissions

The evaluation of formaldehyde emissions was performed on 3 specimens of each sample with dimensions of 200 mm length × 381 mm width × 150 mm thick, according to the American standard ASTM D6007 [31] by MASISA S.A. with a dynamic microchamber (DMC). The results can be compared with the standard TSCA-VI [32].

2.8. Analysis of Data

From the results obtained, an ANOVA was performed where the differences between the averages were accepted at a significance of $p < 0.05$. When the differences between the results of every specimen tested were statistically significant, a multivariance analysis LSD test was applied using Statgraphics Centurion 19 software.

Results of the data analyses are marked in each relevant table.

3. Results

3.1. Evaluation of Accelerated UV Aging

Digital images of the exposed surfaces show noticeable color changes for all exposed samples (Figure 4). Color changes due to accelerated climatic exposure are mainly due to photodegradation reactions of the lignin present in the wood [33], because one of the first wood cell wall polymers degraded by UV radiation is lignin [34].

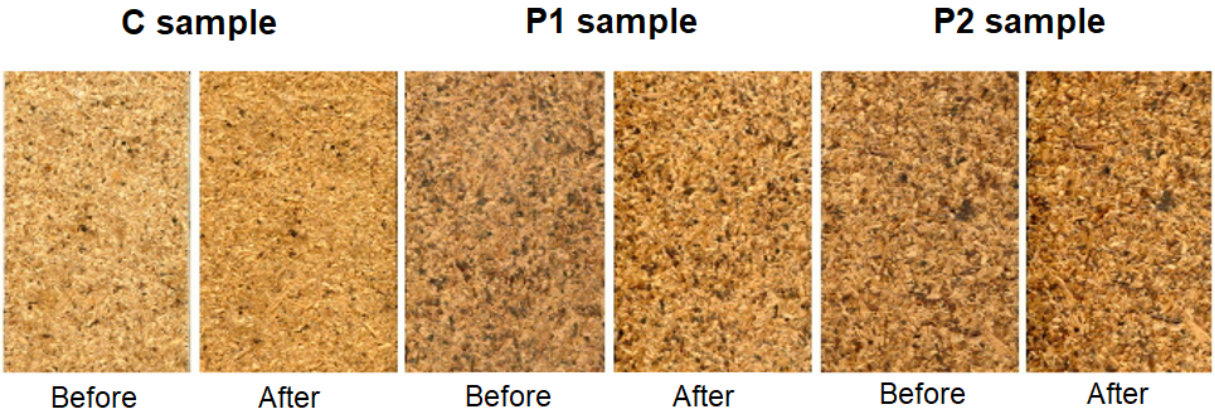

Figure 4. Results of C, P1, and P2 samples before and after exposure to an accelerated UV aging.

The results, shown in Table 3, revealed that specimens of the P2 sample were 50% less susceptible to color change than the control PB sample. There were no statistically significant differences observed between the samples C and P1, nor between the samples P1 and P2.

Table 3. Color change artificial weathering.

ID	ΔE Average
C	3.37 [a]
P1	3.09 [a]
P2	1.68 [b]

Means in columns followed by different letters (a and b) are statistically dissimilar by the LSD test at 95% probability.

3.2. Physical Properties

Physical properties result of all the PB samples under untreated and UV-treated conditions are listed in Table 4.

Table 4. Results of physical properties of all the PB samples under untreated and UV-treated conditions.

ID	Density (kg/m^3)		Moisture Content (%)		Thickness Swelling (%)				Water Absorption (%)			
					24 h		48 h		24 h		48 h	
	Mean	St. Dev.	Mean	St. Dev.	Mean	St. Dev.	Mean	St. Dev.	Mean	St. Dev.	Mean	St. Dev.
C	649 [a]	20.14	4.1 [b]	1.19	23.1 [b]	4.19	24.1 [b]	3.95	91.8 [c]	0.05	97.7 [c]	0.05
P1	718 [c]	51.81	1.4 [a]	0.24	28.6 [c]	3.89	30.6 [c]	3.82	88.9 [bc]	0.09	93.8 [bc]	0.10
P2	724 [c]	50.42	1.7 [a]	0.31	29.2 [c]	3.89	30.4 [c]	3.92	88.4 [bc]	0.12	93.6 [bc]	0.13
C-UV	596 [a]	16.28	8.5 [c]	0.16	14.6 [a]	6.75	15.2 [a]	7.15	79.6 [ab]	14.01	84.0 [ab]	16.32
P1-UV	709 [bc]	55.76	8.0 [c]	0.26	17.9 [a]	2.28	18.7 [ab]	2.43	67.0 [a]	7.29	69.5 [a]	8.84
P2-UV	648 [ab]	39.40	7.7 [c]	0.19	19.1 [ab]	2.14	25.0 [bc]	8.48	82.9 [bc]	6.43	86.8 [bc]	7.36

Means in columns followed by different letters (a, b and c) are statistically dissimilar by the LSD test at 95% probability.

The PB based on bio-wood adhesives showed an increase in density compared to the control panel. The range of increase was between 10.63% and 11.56% for the P1 sample (718 kg/m^3) and the P2 sample (724 kg/m^3), respectively.

The density profile showed differences in the PB samples as shown in Figure 5. The samples P1 and P2 presented a more constant density along the profile than sample C, which showed a clear difference between the surface layers (highest density values) and the middle layer (lowest density values).

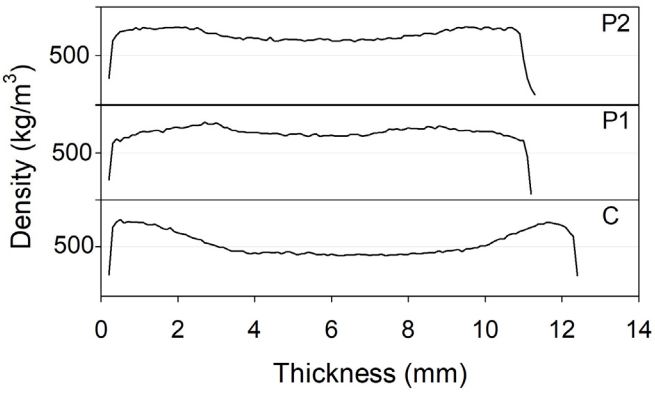

Figure 5. Density profile of each sample.

The difference in density values over the thickness between all the samples tested can be attributed to the different particle sizes used along the PB thickness and, in the case of the control sample, different types of adhesives in the different layers of the PB were also used (for which the characteristics are listed in Table 1). Despite this difference in densities within each board, the average densities per sample are within the range of the commercial standard of 492–800 kg/m^3 [35].

The difference in the thickness of the specimens may be attributed to the moisture content of each one. Table 4 shows that sample C had a higher moisture content than samples P1 and P2, which may have affected their thicknesses.

The samples tested before and after the UV aging showed no statistical differences between samples with the same adhesive. Also, in the case of this investigation, UV aging does not cause changes in the density values of the samples.

Bio-based wood adhesives were manufactured to have a 2% moisture content, so the results obtained are consistent with the initial proposal.

After the UV aging was applied, the samples presented an increase in the moisture content property compared to the untreated samples, which was expected after the samples were subjected to steam and temperature cycling. Fortunately, the results were considered acceptable because they were within the expected range of conditioning.

The thickness swelling (TS) of the samples made of bio-based wood adhesives showed an increase compared to the control panel. The range of increase in 24 h of water immersion was between 23.81% and 26.41% for the P1 sample (with a TS of 28.6%) and the P2 sample (with a TS of 29.2%), respectively.

The range of increase in thickness swelling in 48 h of water immersion, was between 26.97% and 26.14% for the P1 sample (with a TS of 30.6%) and the P2 sample (with a TS of 30.4%), respectively.

This means that the control PB is ~25% more dimensional stable than PB-bbwa. However, the results were considered acceptable given that these PBs still kept a good cohesion after the immersion in water without disassembling [16] and are consistent with the results of the Londoño investigation [8].

After the UV aging was applied, the samples did not present statistically significant differences in thickness swelling at 24 h, but there was a decrease when compared to the untreated samples between 59.91% and 52.52% for the P1 sample (with a TS of 17.9%) and the P2 sample (with a TS of 19.1%), respectively.

After the UV aging was applied, sample P2 presented an increase of 64.47% in thickness swelling at 48 h compared to the control PB sample. On the other hand, there was a decrease when compared to the untreated samples between 63.81% (P1: 18.7%) and 21.77% (P2: 25%).

The results in water absorption of the samples after 24 and 48 h of water immersion did not present statistically significant differences between the bio-based wood adhesive samples and the control PB sample. This indicates that the bio-based wood adhesives are comparable to industrial UF adhesives.

After the UV aging was applied, sample P1 presented a decrease in the water absorption at 24 and 48 h compared to the untreated P1 sample, of between 32.69% and 34.96%, respectively.

All of the PB samples, after 24 and 48 h of water immersion, resisted without disassembling under untreated and UV-treated conditions. Additionally, after UV aging was applied, every sample presented a decrease in thickness swelling and in water absorption properties. This hydrophobic behavior would lead to a better dimensional stability of UV-treated samples in comparison to the untreated samples.

3.3. Mechanical Properties

3.3.1. Stiffness and Strength Bending Properties

PB-bbwas presented a similar behavior to the control PB. These results are shown in Table 5, and in Figures 6 and 7.

Table 5. Results of stiffness and strength bending properties of particleboards.

ID	MOE (MPa)		MOR (MPa)		Density (kg/m^3)		Moisture Content (%)	
	Mean	St. Dev.	Mean	St. Dev.	Mean	St. Dev.	Mean	St. Dev.
C	2394 [a]	530	14.5 [a]	3.1	665	42.2	4.2	0.4
P1	1991 [b]	252	10.6 [b]	1.7	728	45.1	2.3	0.4
P2	2378 [a]	349	11.8 [b]	1.5	763	96.0	2.5	0.5

Means in columns followed by different letters (a and b) are statistically dissimilar by the LSD test at 95% probability.

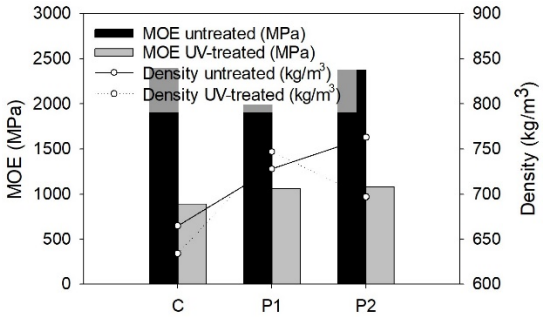

Figure 6. MOE and density results, obtained from the bending test, for each of the UV-treated samples and the untreated samples of the control PB and the PB-bbwas.

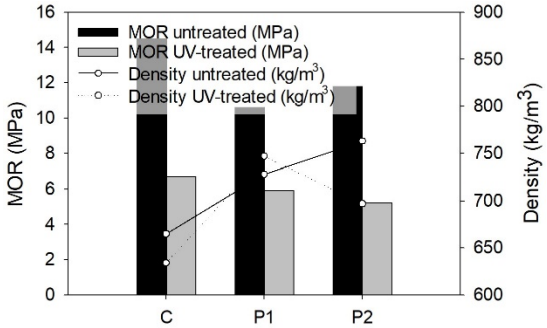

Figure 7. MOR and density results, obtained from the bending test, for each of the UV-treated samples and the untreated samples of the control PB and the PB-bbwas.

The P1 sample (1991 MPa) reached 83.17% of the stiffness (MOE) obtained by the control PB sample (2394 MPa), while the P2 sample (2378 MPa) presented a statistically identical stiffness result to the C sample. The MOE of P1 sample met the EN-312 minimum requirements for type-P2 particleboard of 1800 MPa, while the MOE of C and P2 samples met the minimum requirements for type-P4 particleboard of 2300 MPa.

About the flexural strength of the particleboards, the P1 (10.6 MPa) and P2 (11.8 MPa) samples reached 73.10% and 81.38% of the strength (MOR) obtained by the control PB sample, respectively. The MOR of P1 sample met the EN-312 minimum requirements for type-P1 particleboard of 10.5 MPa, while the MOR of C and P2 samples met the minimum requirements for type-P2 particleboard of 11 MPa.

Flexural stiffness results of the P2 and C samples are consistent with the results obtained in the investigation of PB made of soy flour with urea formaldehyde [17], while flexural strength of the PB-bbwa samples are ~57% higher than the results obtained by the same authors.

The differences in the bending properties results between the PB-bbwa samples can be associated with the difference in the density of each sample, which is consistent with the investigation of the author Umemura [36], who obtained better results of bending stiffness and strength in denser samples than in less dense samples.

After the UV aging was applied, the samples presented a decrease in the stiffness and strength properties compared to the normal samples, and there were no statistically significant differences between the samples subjected to an accelerated UV aging.

By comparing untreated and treated samples, in the case of MOE results, the sample with the lowest decrease was P1 with 88%, while sample C had the highest decrease of 168% and, in the case of MOR results, the sample with the lowest decrease was P1 with 80%, while sample P2 had the highest decrease of 127%. In both cases (MOE and MOR), the PB-bbwa samples demonstrated similar results among themselves, presenting a better flexural stiffness than the control PB sample under UV aging.

The load deflection curves of the bending tests of the samples have evidenced that PB-bbwa untreated samples have decreased the maximum load and maximum deflection; these results are shown in Figure 8.

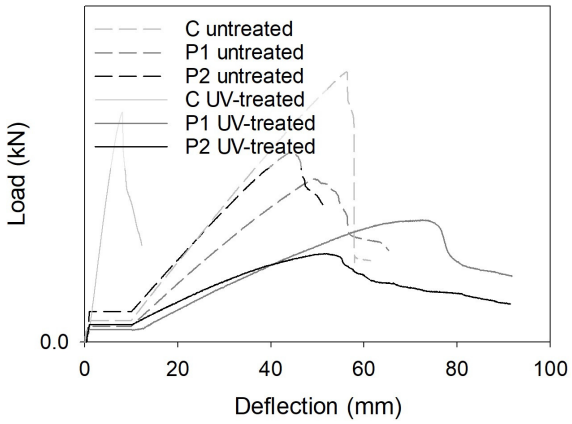

Figure 8. Load deflection curves, obtained from the bending test, for each of the UV-treated samples and the untreated samples of the control PB and the PB-bbwa.

The control PB sample presents a rigid performance, which is a characteristic of UF adhesives, which present a high strength, but a brittle and inelastic performance [37].

PB-bbwa samples showed a slightly more flexible behavior than the control PB sample; this performance can be compared to the study of Nicolao, where samples with more protein content presented a more flexible behavior than the rest of the samples [38].

The bending behavior of each sample can be clearly observed after the UV aging. In Figure 8, it can be seen that the PB-bbwa UV-treated samples showed a flexible behavior, while the control PB sample showed a rigid behavior.

3.3.2. Tensile Strength

PB-bbwas presented a similar behavior in tensile strength (TS) to the control PB. These results are shown in Figure 9.

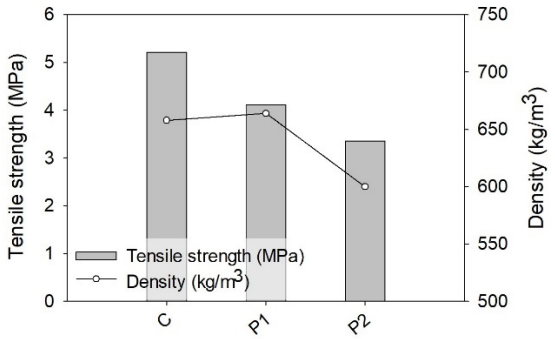

Figure 9. Tensile strength and density results, obtained from the tensile strength test, of each control PB and PB-bbwa sample.

The P1 sample (4.12 MPa) presented a statistically identical TS result to the C sample (5.22 MPa) and of the P2 sample (3.36 MPa), while the P2 sample reached 55.47% of the TS obtained by the control PB sample.

In the tensile test, the specimens are pulled or elongated. The adhesive will elongate first during the test and, after the adhesive cannot elongate any more, the load is transferred to the wood particles [39].

The results obtained can be attributed to the viscosity values of the respective adhesives used. Both the P1 and C samples exhibited statistically similar tensile strength (TS) values, which can be attributed to the adhesives used, as they had very similar viscosity values (Table 1). Conversely, the P2 sample showed a lower TS value, which can be attributed to the adhesive with the lowest viscosity (Table 1). Similar viscosity–tensile strength relationships were observed between the control PB and PB-bbwa density–tensile strengths. These observed relationships can be explained by the fact that the viscosity of a liquid is influenced by its density [40]. As Marinho concluded, density and homogeneity in the profile of the adhesive positively impact the mechanical properties of the tested samples [41].

3.3.3. Internal Bond Strength

PB-bbwa presented a similar behavior in internal bond strength (IB) to the control PB. These results are shown in Figure 10.

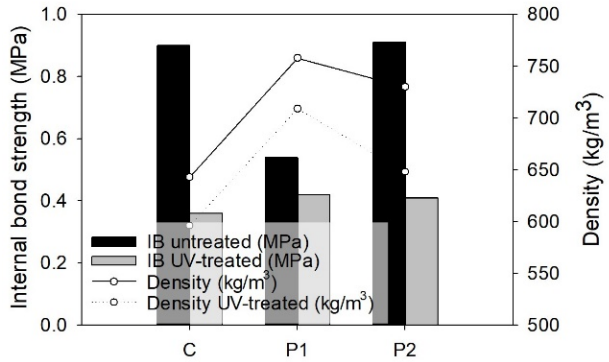

Figure 10. Internal bond strength and density results, obtained from the internal bond strength test, for each of the UV-treated samples and untreated samples of the control PB and PB-bbwas.

The P1 sample (0.54 MPa) reached 60% of the IB obtained by the control PB sample (0.90 MPa), while the P2 sample (0.91 MPa) presented a statistically identical IB result to the control PB sample. The IB of P1 sample met the EN-312 minimum requirements for type-P5 particleboard of 0.45 MPa, while the MOR of C and P2 sample met the minimum requirements for type-P7 particleboard of 0.75 MPa.

After the UV aging was applied, the samples presented a decrease in the IB property compared to the untreated samples, and there were no statistically significant differences between the samples subjected to an accelerated UV aging.

By comparing untreated and treated samples, the sample with the lowest decrease was P1 (0.42 MPa) with 29%, while sample C (0.36 MPa) had the highest decrease of 151%. These results indicated that the PB-bbwas present better internal bond strength results than the control PB under accelerated UV aging.

These results can be attributed to the capacity of bio-based wood adhesives as a protection coating against aging from UV exposure, like the modified recycled palm oil adhesive used by some researchers [40].

3.3.4. Janka Hardness

The PB-bbwa had an increased Janka hardness (JH) compared to the control PB (Figure 11).

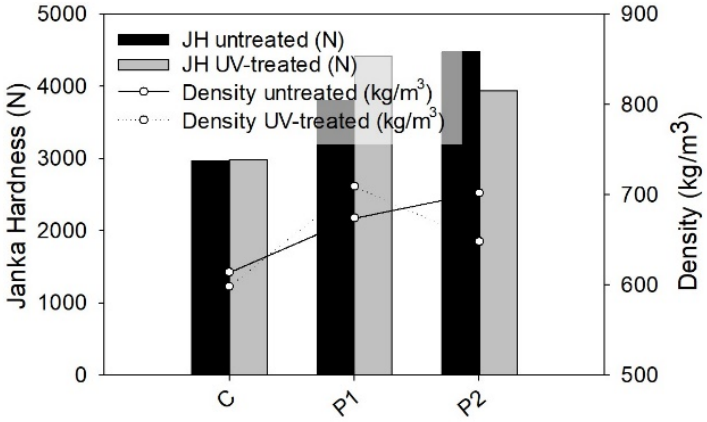

Figure 11. Hardness and density, obtained from the Janka hardness test, for each of the UV-treated samples and the untreated samples of the control PB and PB-bbwas.

The JH of the untreated samples P1 (3801 N) and P2 (4482 N) increased by 27.81% and 50.71%, respectively, compared to the control PB sample (2974 N).

The results obtained can be attributed to the PB density values, because when higher JH results were obtained, the PB density was higher. The same analysis was made by the authors who studied the properties of cotton carpel-based particleboards [41].

After the UV aging was applied, the Janka hardness of the UV-treated samples P1 (4424 N) and P2 (3943 N) increased by 48.06% and 31.95%, respectively, compared to the control PB sample (2988 N). Additionally, there were statistically significant differences between the PB-bbwa samples and the control PB sample, all of which were subjected to accelerated UV aging.

By comparing untreated and UV-treated samples, the sample with the highest increase was P1 with 16.39%, while sample P2 decreased by 13.67%. These results indicate that the sample made with P1 bio-based wood adhesive presents better JH results than the rest of the samples under UV aging.

These results can also be attributed to the ability of bio-based wood adhesives to function as a protective coating against aging from UV exposure, as mentioned in Section 3.3.3 in the study by Suwan et al., 2020 [42]. However, in this instance, the coating not only provided protection but also enhanced the hardness property of the particleboards.

3.4. Thermal Properties

PB-bbwas, in edgewise position (EW), presented a similar behavior in thermal conductivity to the control PB, while in flatwise position (FW) presented statistically identical results to the control PB. These results are shown in Figure 12.

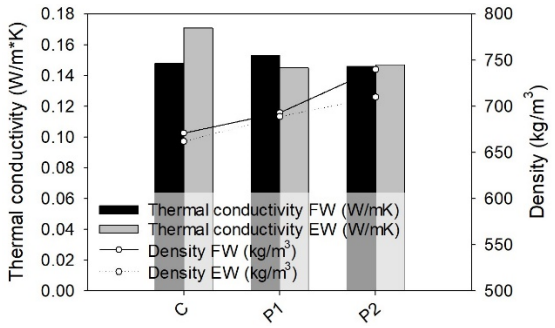

Figure 12. Thermal conductivity and density, obtained from the thermal conductivity test, for each of the flatwise and edgewise positions of the control PB and PB-bbwas.

In the EW position, both the P1 and P2 samples (0.14 W/mK) demonstrated the same results, exhibiting a decrease of approximately 11.76% in the thermal conductivity compared to the control PB sample (0.17 W/mK), which was expected considering that protein-based adhesives present lower thermal conductivity values than urea formaldehyde adhesives [43,44].These results are a good indicator, since PB-bbwa in the edgewise position would be a more insulating material than commercial urea-formaldehyde-based particleboards [40].

3.5. Evaluation of Formaldehyde Emissions

Wood itself emits different compounds, including formaldehyde, which can vary depending on the wood species [45]. In the case of the evaluation of formaldehyde emissions in this research, the results, which are presented in Table 6, were obtained for two cases: the control PB sample, in which the measurement yielded formaldehyde emissions coming from the wood and in addition to the UF adhesive; and a second case corresponding to the PB-bbwa samples, in which the measurement yielded formaldehyde emissions only from the wood, since the bio-based wood adhesives used in this research are free of this compound.

Table 6. Formaldehyde emissions at 2 h and 7 days for PB.

ID	2 h (ppm)	7 Days (ppm)
C	0.068	0.042
P1	0.010	0.004
P2	0.010	0.005

From the results obtained, it could be seen that when bio-based wood adhesives are used, formaldehyde emissions from particleboard are significantly reduced compared to the control PB sample, leaving only the volatile organic compounds (VOCs) from the wood itself to emit formaldehyde [45].

At 2 h, the formaldehyde emissions of the samples P1 (0.01 ppm) and P2 (0.01 ppm), decreased by 85.29% compared to the control PB sample (0.068 ppm), while in 7 days, the emissions of the samples P1 (0.004 ppm) and P2 (0.005 ppm), decreased by 88.10% compared to the control PB sample (0.042 ppm).

The results obtained met the Toxic Substances Control Act (TSCA-VI) [32] requirements of formaldehyde emissions, namely 0.09 ppm of formaldehyde in PB. Some authors also obtained a considerable decrease in their PB-bbwa's formaldehyde emissions, and with their results they validated the use of these adhesives in the manufacturing of PBs at a laboratory scale [16].

4. Conclusions

The tests performed evidenced that particleboards with bio-based wood adhesives showed comparable performance against the control PBs made with urea-formaldehyde and satisfied some international standard requirements.

PB-bbwas presented an increase in density values of around 10% compared to the control PB. After water immersion, PB-bbwas presented similar results in thickness swelling and statistically identical results in water absorption in comparison to commercial PB; additionally, they kept a good cohesion without disassembling.

P1 sample reached 83% and 73% of the control PB sample flexural stiffness and strength, respectively, while P2 sample presented a statistically identical flexural stiffness to the control PB sample, and reached 81% of its flexural strength.

The P1 sample, presented a statistically identical tensile strength to the control PB sample, and reached 60% of the control PB sample internal bond strength and had an improved Janka hardness of 28% compared to the control PB sample. On the other hand, the P2 sample reached 55% of the control PB sample's tensile strength, presented statistically identical IB results to the control PB sample, and an improved Janka hardness of 51% compared to the control PB sample.

PB-bbwas presented statistically identical thermal conductivity in a flatwise position to the control PB and a decrease of 12% the same property in edgewise position. They also decreased the formaldehyde emissions in 7 days by 88% compared to the control PB results.

PB-bbwas under accelerated UV aging presented higher density values, hydrophobic behavior under water immersion, and similar flexural behavior and hardness compared to the control PB. Furthermore, the UV-treated samples presented similar hardness results compared to the untreated samples.

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