Shelf-Life Assessment of Canola Protein Bio-Adhesive

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The storage stability of binders before their application is a crucial factor in the wood panel industry, as it impacts the mechanical properties, quality control, economic efficiency, and market competitiveness of the final products. In the present study, the long-term stability of two canola protein isolate (CPI) and two canola meal (CM) adhesive variants was investigated. The protein-based adhesives were prepared and tested on one-layer particleboards after one week, one month, two months, three months, and four months of storage of the formulations. Results indicate that the CPI-based outperformed the CM-based variants in terms of internal bonding strength (IB), modulus of rupture (MOR), and modulus of elasticity (MOE) due to the higher protein concentration of the CPI over the CM. While the IB strength of the CM-bonded particleboards was lower than the EN 319 requirement after the first four weeks of storage (0.34 N/mm^2 and 0.29 N/mm^2 for nitrite and bisulfate-crosslinked respectively), that of the CPI-bonded was still superior to the EN 319 after four months (0.44 N/mm^2 and 0.3 N/mm^2 for nitrite and bisulfate-crosslinked respectively). This indicates that the nitrite-crosslinked variants had a more robust chemical formulation, leading to stronger and more durable bonds.

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INTRODUCTION

Particleboard is a major wood-based composite material that has been gaining popularity for many years, finding use in various applications (Pizzi *et al.* 2020). In 2022, global particleboard production reached 110.25 million m^3, marking a 13.7% increase over 2020 levels (FAO 2023), with Europe and Asia leading in production. As a result, the demand for adhesives has also increased. Currently, the wood-based industry relies heavily on synthetic resins derived from petroleum-based components such as urea, phenol, and melamine (Mantanis *et al.* 2018; Ostendorf *et al.* 2021a; Dorieh *et al.* 2022a,b). Although these synthetic binders offer enhanced efficiency and performance, their environmental and health impacts, coupled with their unsustainable nature, have motivated a search for cleaner alternatives (Li *et al.* 2012; Tene Tayo *et al.* 2022). Consequently, the development of bio-based adhesives has become a critical objective for a sustainable production of particleboards (Arbenz and Avérous 2015; Xu *et al.* 2020; Hussin *et al.* 2022).

This ongoing transition towards a more sustainable and resilient bioeconomy is essential for decarbonizing the sector and adopting a circular development model (Cárdenas-Oscanoa *et al.* 2024a,b; Tene Tayo *et al.* 2024). By reshaping linear industrial value chains to minimize pollution and waste generation, this shift supports greater sustainability and inclusivity, effectively addressing climate change and reducing reliance on fossil-based materials (Antov *et al.* 2023). Over the past decades, plant proteins have emerged as a reliable alternative for adhesives development, with soy receiving the most attention to date (Vnučec *et al.* 2017; Li *et al.* 2022a,b; Chen *et al.* 2023). Despite the potential of soy as raw material for the development of green adhesive systems, there is a growing recognition of the need to diversify protein sources for bio-adhesives (Solt *et al.* 2019; Barzegar *et al.* 2020, 2022; Dunky 2021; Frihart 2023; Kallakas *et al.* 2024) to ensure the long-term production and supply of protein-based bio-adhesives. This diversification is crucial for enhancing the resilience of the wood-based panel industry by reducing dependence on a single protein source (Tene Tayo *et al.* 2024).

Recent agricultural development has boosted the production of canola worldwide. As result, canola is to date ranked as second most abundant oilseed after soy (Goyal *et al.* 2021). Cultivated primarily for its oil, canola processing generates substantial by-products, which have been relegated to low-value applications such as animal feed or fertilizer (Manamperi *et al.* 2010; Wang *et al.* 2014). Thus far, the potential of defatted canola as an industrial raw material remains underutilized (Adhikari *et al.* 2017; Li *et al.* 2017a). Exploring high-value applications of canola protein, such as in bio-adhesive production for wood composites, will significantly enhance the economic viability of the canola oil industry (Manamperi *et al.* 2010). Only a small number of studies have explored its use in bio-adhesives, employing either canola protein isolates (Wang *et al.* 2014; Bandara *et al.* 2017; Li *et al.* 2017a), canola flour (Yang *et al.* 2010, 2011; Ostendorf *et al.* 2021a,b). Among these, adhesives formulations based on the protein isolates have demonstrated superior bonding strength compared to those based on crude canola meal. However, using canola meal instead of isolated proteins offers a more practical alternative, as it circumvents the high costs and low yields associated with protein extraction (Elstner and Stein 1982). This approach could pave the way for more accessible and sustainable bio-adhesive solutions while leveraging the existing by-products of canola processing. Therefore, further research is needed to optimise the performance of meal-based adhesives to match the strength and reliability of isolate-based formulations, ensuring their industrial viability

Unfortunately, research and development efforts on adhesives derived from natural resources have primarily focused on creating the adhesive products themselves, overlooking the substantial quantities required if the production of wood-based panels were to rely exclusively on these natural adhesives (Dunky 2021). Moreover, the industrial application of these innovative formaldehyde-free adhesives is often overlooked, making their upscaling even more challenging. In their thorough evaluation of innovative adhesive systems for wood-based panels, Solt *et al.* (2019) compared synthetic and renewable-based adhesives, focusing on assessing the individual adhesive systems based on selected technological parameters relevant to wood-particleboard production. They concluded that numerous reports in the literature demonstrate favourable board properties. Regrettably, these boards are frequently manufactured using excessively long press times, far beyond any industrial norms and even those observed in niche products and laboratory conditions (Dunky 2021). Additionally, due to the high susceptibility of protein-based bio-adhesives to microbial attack, which can significantly compromise their service life and performance, protein-based adhesives are typically tested fresh. This poses a problem for upscaling, as most wood-based industries prefer ready-to-use products, as onsite production requires extra investment in production equipment. Some scholars have attempted to address this issue through different strategies such as incorporation of silver and ZnO nanoparticles to inhibit microbial growth and effectively improve the storage of the adhesive (He *et al.* 2020). Similarly, chitosan coatings, known for their antimicrobial properties, have also been successful in preserving bio-adhesives (Saleh *et al.* 2021). A recent study highlighted a soy protein-based adhesive that demonstrated remarkable resistance to mildew, with its stability against mould growth improving dramatically from 1 day to over 15 days (Xu *et al.* 2021). This enhanced resistance was attributed to a synergistic effect within the adhesive structure, involving covalent, hydrogen, and ionic bonds, which reinforced its overall stability. However, while tailoring bio-adhesive formulations or incorporating preservatives, a key consideration remains ensuring that the adhesive's bonding performance and mechanical properties are not adversely affected; and the delicate balance between microbial resistance and adhesive performance still requires extensive investigation, particularly to optimize the formulation for large-scale applications (Yue *et al.* 2023). Further studies in this area will contribute to improving the stability of protein-based bio-adhesives while maintaining their bonding performance.

Producing bio-adhesives onsite in the wood-based panel industry involves overcoming significant technical, economic, and regulatory challenges. While the potential environmental and health benefits of bio-adhesives make them an attractive alternative to synthetic adhesives, the complexity of production, high initial and operational costs, and stringent regulatory requirements must be carefully managed. Achieving consistent quality and performance comparable to that of traditional synthetic adhesives is crucial for widespread adoption in the industry.

Although collaboration with research institutions, investment in technology, and strategic planning can help address these challenges and make onsite bio-adhesive production viable, obtaining a ready-to-use, storage-stable flowable adhesive formulation remains a better option, as it limits the investment related to production. Because natural adhesives may have shorter shelf lives compared to synthetic adhesives, ensuring stability over time and under varying storage conditions is a challenge. Hence, understanding the behaviour of newly developed adhesive systems is paramount for their industrial application, and thorough investigations of their responses to various production parameters in the context of particleboard manufacturing are essential. Therefore, this study aims to explore the impact of the adhesive formulations and their storage time on the mechanical properties of one-layer particleboards. Given the already known low water resistance of protein-based adhesives, the canola-bonded particleboards produced and tested in the present study were designed for indoor application in dry conditions (P2 grade).

EXPERIMENTAL

Materials

The canola protein (Puratein® G) isolate (CPI) with a protein concentration of 90% was purchased from Merit functional Foods, Winnipeg, Canada. The canola meal (CM) was offered by Kleeschulte GmbH & Co. KG (Büren, Germany). This by-product of the canola oil manufacturing process arrived in the form of pellets. Sodium dodecyl sulfate

(SDS), urea, sodium bisulfate (92%), sodium chloride, and sodium nitrite (99%) were sourced from VWR International in Darmstadt, Hesse, Germany. Sodium bisulfate, an acidic salt produced by partially neutralizing sulfuric acid with sodium hydroxide or sodium chloride, appears as a dry granular substance with hygroscopic properties. In contrast, sodium nitrite, an inorganic compound with the chemical formula $NaNO_2$, presents as a white to slightly yellowish crystalline powder that readily dissolves in water and exhibits hygroscopic characteristics. The Gelatine (180 Bloom) was obtained from Carl Roth GmbH + Co KG in Karlsruhe, Germany. The hydrophobic agent, SASOL Wax Pro 18A, with a solids content of 60%, was acquired from SASOL Wax GmbH in Hamburg, Germany. The industrial wood particle material obtained from a residual process was supplied by Pfleiderer in Arnsberg, North Rhine-Westphalia, Germany.

Adhesive Preparation

The preparation of the canola protein-based adhesives used in the present study followed the steps described in Tene Tayo *et al.* (2024). To begin, a gel mixture comprising gelatine, urea, SDS, and water in the proportions of 25:24.5:0.5:50, respectively, was prepared. After ensuring complete dissolution of the gelatine and obtaining a homogeneous slurry, the mixture was conditioned in an oven set at 25 °C for 72 h before being utilized for binder preparation. Subsequently, the necessary amount of a 1 mol solution of NaOH was added to the gel mixture, along with sodium chloride (NaCl) and either sodium bisulfate ($NaHSO_4$) or sodium nitrite ($NaNO_2$) (Proportions are outlined in Table 1). Sodium chloride was therefore employed to enhance protein solubility, thereby contributing to the solid content of the adhesive (Tene Tayo *et al.* 2024).

Table 1. Adhesive Formulation

Components	Proportions (%wt)	
	CPI-based	CM-based
Canola	25.6	14.16
Gel mixture	51.2	34.62
Sodium chloride	3.07	1.88
Sodium bisulfate/sodium nitrite	3.07	1.88
Sodium hydroxide*	17.05	47.44
Total solid content of the adhesive (%)	55	35

* The sodium hydroxide is a 1 mol solution

The CPI/CM was then gradually added while stirring with an RW 20 laboratory stirrer from IKA®-Werke GmbH & Co. KG, Staufen im Breisgau, Germany, rotating at 10,000 rpm. To ensure proper denaturation of the canola protein and expose the active groups of the protein chain, the pH of the slurry was adjusted to 11 ±0.2. The solid content of the resulting binder was 55%. Alkaline denaturation of the proteins caused by NaOH is necessary for unfolding, exposing hydrophobic groups, and increasing the surface area available for interaction with other molecules. This unfolding is essential for enhancing the adhesive qualities of the protein by increasing its ability to form strong bonds.

The effect of heat treatment on the performance of the binder formulations was previously done by incubating at 60 °C for 30, 45, and 60 min using the IKA® LR 1000 modular laboratory reactor from IKA Werke GmbH & Co. KG, rotating at 120 rpm. Results showed that heat treatment has a positive effect on the nitrite-crosslinked variants, but negatively affects the bisulfate-crosslinked adhesive formulations (Tene Tayo *et al.* 2024).

Moreover, this treatment helps improve the viscosity of the bio adhesive formulations, especially for those containing nitrite. Consequently, in the present study, the bisulfate-crosslinked adhesive variants were not subjected to heat treatment and labelled CPI-B-0 and CM-B-0, while nitrite-crosslinked variant was incubated for 60 min at 60 °C and labelled CPI-N-60 and MC-B-0. They were kept sealed at ambient temperature (about 20 °C) and allowed to age before being used to produce one-layer particleboards after one week, one month, two, three and four months of storage.

Fourier Transformed Infrared Spectroscopy (FTIR)

Fourier transformed infrared spectroscopy (FTIR) was used to characterize the crosslinking mechanism and the aging behaviour of the adhesives produced from canola meal and canola protein. The samples were scanned using an Alpha spectrometer (Bruker, Germany). The FTIR spectra of every sample were recorded consecutively in transmission mode in the range of 400 to 4000 cm^{-1} with a spectral resolution of 4 cm^{-1}. Collected IR spectra were processed and analysed with Origin 2017 software (OriginLab Corporation, MA, USA).

Particleboards Productions at Lab Scale

The one-layer particleboards were produced in the Biotechnikum laboratory of the Burckhardt Institute, University of Goettingen, Germany. The wood chips were beforehand dried (overnight at 100 °C) to about three percent moisture content using a Memmert UN45 universal oven. Precise amounts of wood chips and resin were weighed. A resin load of 10% based on the oven-dried wood material was applied onto the wood particles in a rotating blending drum using an air-pressure atomizer nozzle from Düsen-Schlick GmbH, Coburg, Germany. The hydrophobic agent (1%) was mixed with the adhesive prior to application. The boards were preformed using a 0.32 m x 0.42 m mat former. Hot-pressing was conducted using a semi-automatic single-opening Hydraulic Lab hot-press (Siempelkamp Hydraulic Lab Press A 308/1988). The final thickness was adjusted to 14 mm using two stop control bars placed between the pressing plates. A press time factor of 12 s/mm was used for the boards bonded with the CPI-based adhesive variants. Due to the lower solid content of the CM-based formulation and hence, the higher moisture content after adhesive application, a press time factor of 26 s/mm was used. Detailed production parameters are outlined in Table 2.

Table 2. Particleboard Production Parameters

Board Type	One-Layer Particleboard
Target density (kg/m³)	640 kg/m³
Board thickness (mm)	14 mm
Binder formulations	CPI-B-0; CPI-N-60; CM-B-0; CM-N-60
Binder content (%) *	10
Press temperature (° C)	210
Hydrowax (%) *	1
Boards per variant	4

* The binder and the hydrowax content are based on the oven-dried amount of wood particle material used.

Each treatment involved the production of four boards (repetitions). After production, the boards were conditioned at room temperature for 24 hours, followed by trimming to remove edge effects and sanding on both sides using a wide-belt sanding machine (Felder FW 950 C from Felder Group, Hall In Tirol, Austria) before testing.

Testing the Mechanical Properties of the Produced Particleboards

The mechanical properties of the produced particleboards were then assessed through various tests. The internal bonding strength (IB) was evaluated according to EN 319 (1993), while the modulus of rupture (MOR) and modulus of elasticity (MOE) were determined in accordance with EN 310 (1993). These tests were conducted using a ZWICK/ROELL universal testing machine (type 10) from MFC Sensortechnik GmbH, Wuppertal, Germany. For the MOR and MOE tests, five specimens measuring 50 by 400 mm were taken from each produced board. Similarly, for the IB test, five test pieces were prepared from each produced board. Here, the selection process of the specimen was based on the density. The boards were cut into 50 by 50 mm pieces. The weight and dimensions were measured and the density of each calculated. Sample with density value closer to the board’s target density were selected for the IB test.

Data Analysis

The data analysis was performed for the mechanical properties of particleboards. An analysis of variance (ANOVA) ($p < 0.05$) was conducted to test the significancy of the influence of the factors on the mechanical properties of the particleboards followed by a pair wise mean comparison when necessary.

RESULTS AND DISCUSSION

The crosslinking effect of nitrite and bisulfate was found to introduce specific structural modifications to the protein backbone, enhancing the adhesive’s functional and mechanical properties (Fig. 1a). The increased absorbance around the 3100 to 3600 cm^{-1} region (O-H/N-H stretching) indicates formation of hydrogen bonding and stretching vibrations of hydroxyl (O-H) and amine (N-H) groups. The ionic nature of bisulfate favors the formation of additional polar groups such as sulfonate or sulfate, which can connect with other functional groups in the adhesive matrix, forming hydrogen bonds (Zhu *et al.* 2016). Very little changes were observed for the C-H stretching (2800 to 3000 cm^{-1}), suggesting that crosslinking with nitrite or bisulfate did not significantly disrupt the aliphatic components of the protein. A noticeable change in intensity and a slight shift for the bisulfate- and nitrite-treated samples compared to the reference around the amide I (1600 to 1700 cm^{-1}) and amide II (1500 to 1600 cm^{-1}) bands was observed. The amine I primarily arises from C=O stretching vibrations of the peptide bonds, while amide II is associated with N-H bending and C-N stretching vibrations. Alterations in these bands indicate changes in the secondary structure of proteins. For nitrite-treated adhesive variants, changes in these regions suggest the formation of covalent bonds between nitrite and protein functional groups, such as nitroso crosslinks (R-N=O) or dityrosine bonds formed *via* oxidative coupling of tyrosine residues (Deng 2006; Yeo *et al.* 2008). For bisulfate-treated adhesives, increased intensity may arise from ionic crosslinking between sulfate/sulfonate groups and amine functionalities on the protein, stabilizing the structure and introducing stronger ionic interactions (Zhang *et al.* 2019). A significant increase in absorbance was observed for sulfur-containing groups (1000 to 1300 cm^{-1}), which is typical for a bisulfate-treated adhesive. This region corresponds to the S=O stretching vibrations of sulfate or sulfonate groups. The higher absorbance for bisulfate-treated samples confirms the incorporation of sulfate functional groups into the adhesive matrix (Chen *et al.* 2015). The bisulfate crosslinking introduced ionic interactions and stabilized the adhesive by interacting with protein functional groups (*e.g.*, amine and hydroxyl groups), forming a denser ionic crosslinking network. The formation of new bonds and functional groups such as nitroso groups (R-N=O) through reactions with amine group of the protein chain, the dityrosine bonds formed *via* oxidative coupling of tyrosine residues and, secondary and tertiary amines created *via* nitrite-induced modifications of protein structures led to formation of covalent crosslinking, introducing dityrosine bonds and nitroso groups, which helped enhance the mechanical strength of the nitrite-treated adhesive variants. For the bisulfate-treated variants, the formation of sulfonate ($R\text{-}SO_3^-$) or sulfate groups improved ionic interaction between sulfate groups and amine or hydroxyl functionalities. The alterations in the fingerprint region of the bisulfate-treated samples indicate changes in the adhesive’s polymeric structure, likely due to bisulfate-induced ionic interactions and potential esterification or dehydration reactions facilitated by heat and acidic conditions (Hale 2013).

In canola, FTIR spectra distinctly reveal vibrations from specific amino acid side chains, facilitating the pinpointing of site-specific protein modifications. Particularly, significant absorbance is observed for amino acids such as Tyr, His, Arginine (Arg), Asparagine (Asn), Glutamine (Gln), and Lysine (Lys) compared to others (Bandara and Wu 2018). The band around 1157 cm^{-1} is attributed to the stretching of the C-O bonds of aliphatic esters (Chen *et al.* 2015; Vlachos *et al.* 2006; Yang and Irudayaraj 2000). Adhesives treated with sodium nitrite exhibited increased intensity after 4 months of exposure compared to 1 month and 1 week (Fig. 1b). However, this effect was not observed in adhesives treated with sodium bisulfate (Fig. 1c). Similar to Tyr, notable absorbance intensities were observed for the –NH vibration at 1240 cm^{-1} and the –CN vibrations at 1440 cm^{-1}. The absorbance intensities of Tyr ring –OH group vibrations at 1518 cm^{-1} and 1602 cm^{-1} were also significant, particularly in protein isolated adhesives compared to canola meal, possibly indicating initiated crosslinking and reduced -OH ring vibration residue (Bandara and Wu 2018). The amide bands, characteristic features of proteins, observed at 1623, 1531, and 1238 cm^{-1} were assigned to amide I (C=O stretching), amide II (N-H bending, primary bands of peptide linkage), and amide III (C-N stretching and N H bending) (Barzegar *et al.* 2020). These peaks were more intense in protein isolate spectra (Fig. 1b, 1c). The prominent absorption band at 1743 cm^{-1} is attributed to the C=O stretching of aliphatic esters. Its intensity was stronger in protein isolate adhesives compared to canola meal, especially after 4 months, with the exception of canola meal treated with sodium bisulfate.

The strong bands around 2922 and 2852 cm^{-1} are attributed to the asymmetrical and symmetrical C-H stretching vibrations of CH_2 groups (Li *et al.* 2017b; Yang and Irudayaraj 2000). Interestingly, these peaks appeared more pronounced in canola meal adhesives compared to protein isolate adhesives. A peak observed at 3300 cm^{-1}, indicating the presence of free and bound O-H and N-H groups, suggests a higher water content in canola meal adhesives (sodium bisulfate and sodium nitrite) due to their lower solid content (35% vs. 55% for protein isolate adhesives). As anticipated, moisture levels decrease in all adhesives after 4 months.

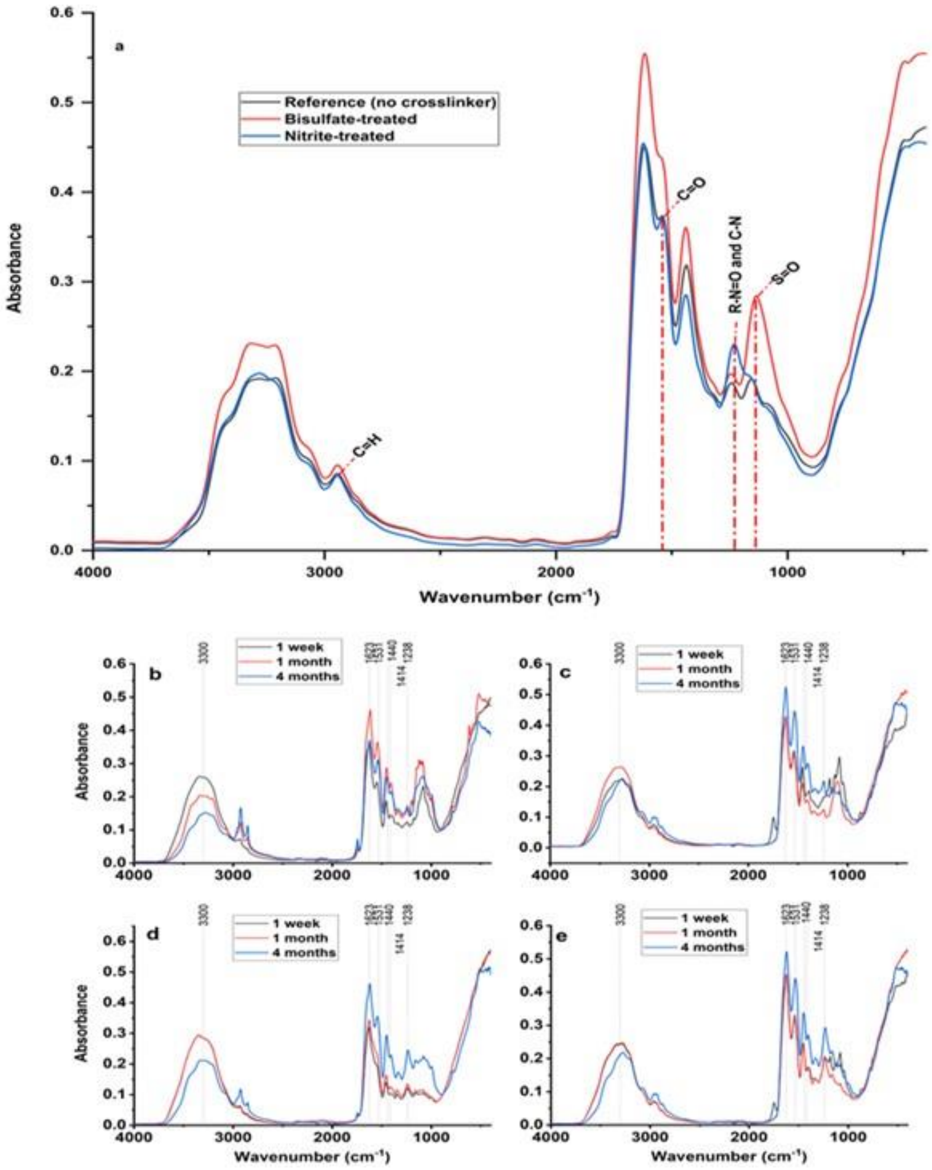

Fig. 1. FTIR spectra of the different adhesive formulations. a) Crosslinking effect of nitrite and bisulfate; and effect of storage of the different adhesive formulations: b) CPI-B-0; c) CPI-N-60; d) CM-B-0; e) CM-N-60

Rheological Properties of the Binder Formulations

The viscosity of a wood adhesive plays a crucial role in this process as well, affecting the penetration behaviour of the adhesive into the wood surface (Scheikl and Dunky 1998). Achieving a sufficiently strong bond requires proper penetration of the adhesive depth into the wood surface, enabling intimate molecular contact with the substrate (Cheng and Sun 2006). While a low viscosity leads to excessive penetration, hindering the formation of an adhesive layer between wood particles, very high viscosity makes it difficult to spray the adhesive and results in uneven distribution, thereby affecting panel bonding strength (Tene Tayo 2024). A decrease in viscosity of the adhesives over time was observed (Fig. 2). The bisulfate-crosslinked variants were found to be more affected by the storage than the nitrite-crosslinked ones. Indeed, while reductions of about 53% and 59% were recorded after 1 month and four months of storage respectively for the

CPI-B-0 variant, only 33% and 48% reduction were obtained with the CPI-N-60 for the same periods of time. Also, the viscosity of the CPI-based variant was much more affected over time than that of their CM-based counterparts. Only a 33% and 27% reduction could be observed from the CM-B-0 and CM-N-60, respectively, after a period of one month. The change in the viscosity of the canola-based adhesive formulations over time is likely due to an oxidation process occurring during storage. Exposure to oxygen, especially in the presence of heat or light, can cause the oxidation of the binder's polymer chains, which leads to the formation of smaller molecular fragments. As these larger polymer chains break down into smaller fragments, the overall molecular weight of the binder decreases. Lower molecular weight results in reduced viscosity because the smaller molecules flow more easily compared to the original, larger polymer chains. The higher oxidation rate and hence the higher reduction in the viscosity of the CPI-based adhesives can be attributed to the presence of air bubbles trapped in the adhesive during preparation (Tene Tayo 2024). Furthermore, hydrolytic reactions due to the presence of water can cleave ester bonds or other susceptible linkages within the polymer, breaking down the structure. This process can also lead to the formation of acidic by-products, contributing to the decrease in pH, which was actually observed as the pH of the adhesives dropped to 9 after four months. As observed, the oxidation process was much obvious within the first four weeks of storage, resulting in a substantial reduction in the viscosity. After this period, the oxidation rate decreased. Moreover, the accumulation of these acidic compounds resulting from the abovementioned oxidation led to a lower pH of the binder formulation (the pH dropped from 11 to 9 after 7 months), as well as a slightly higher solid content (from 55% to 57%).

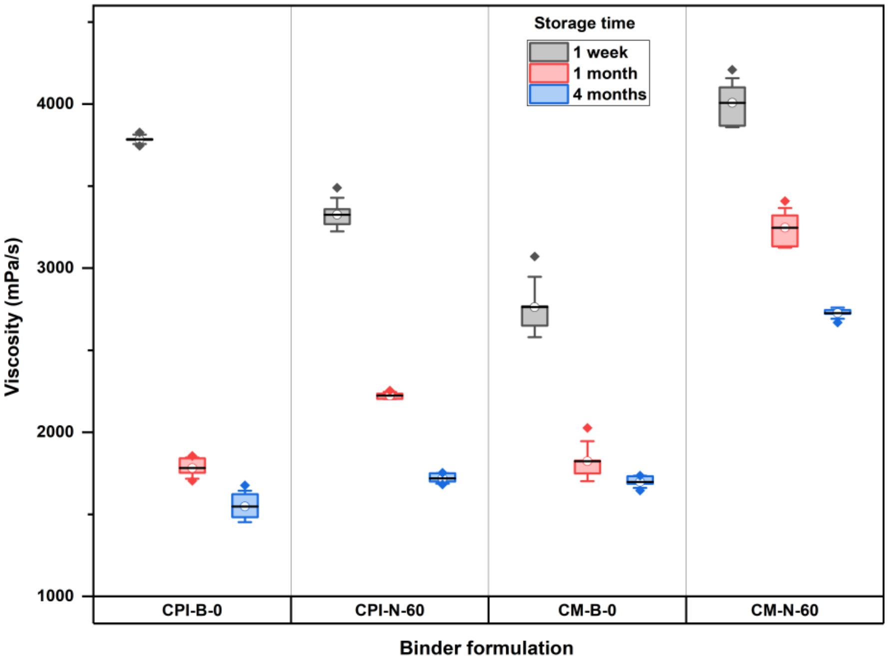

Fig. 2. Apparent viscosity of the different binder formulation as a function of storage time

Mechanical Properties of the One-Layer Particleboards

The performance of the adhesive system used in the production determines the strength and the stability of the particleboard. In the present study, the effect of the binder formulation, protein type (protein isolate or defatted meal), and that of the storage time were investigated to understand their effect on the performance of the developed protein adhesive. The recorded internal bonding (IB) values of the one-layer particleboards provide an insight into the performance and durability of the binder formulation over time. Because of the difference in the press time used (3 minutes for the CPI-based binder formulations versus 7 minutes for the MC-based), the results were not comparable and will be discussed separately.

The results depicted in Fig. 3, show that the nitrite-crosslinked variants were superior to the bisulfate-crosslinked ones. Irrespective of the storage time, CPI-N-60 variants were significantly superior to the CPI-B-0 ($p \leq 0.001$). The superiority of the nitrite treated variants was expected as the same was reported in a previous study where nitrite was observed to be a better crosslinking agent than bisulfate (Tene Tayo 2024). Feng *et al.* (2016) also reported that addition of nitrite to protein leads to increased disulfide bonding, which thereby improves the binding property of the adhesive. The mean IB values of 0.36 and 0.53 N/mm^2 were recorded for the CPI-B-0 and the CPI-N-60 respectively. This represents an increment of over 47%. The BS was as well significantly affected, although with a much lower incremental amount (7%). Although the recorded BS values remained lower than the EN 310 standard, it is worth reminding that the MOR of one-layer particleboards is expected to be lower than that of the three-layers boards that have their surface layer densified by the use of smaller wood particles in the surface layer. This substantially improves the load bearing capacity of the three-layer boards over the single layer.

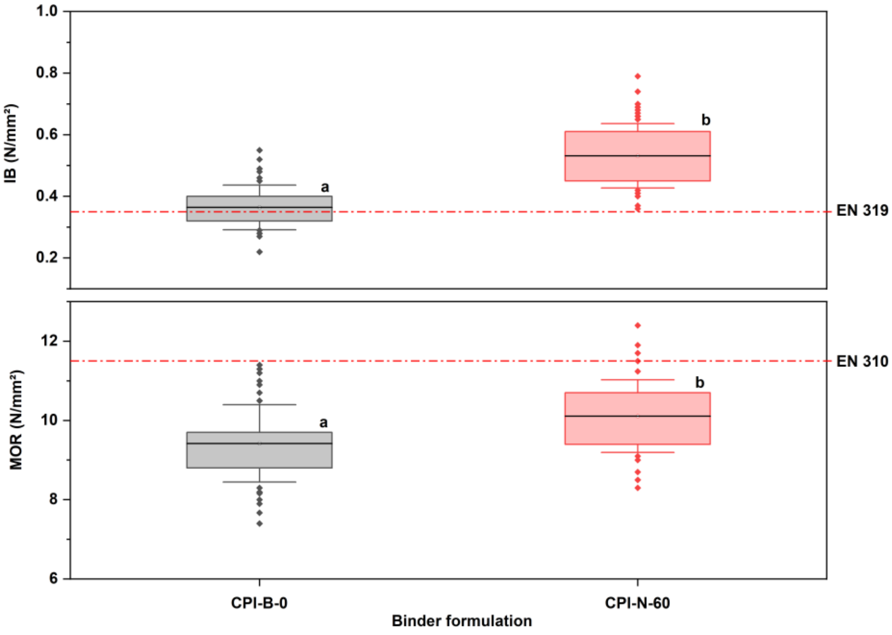

Fig. 3. Effect of crosslinker type on the CPI-based adhesive formulation. Box plots (25th quartile, mean and 75th quartile) and whiskers (1 × standard difference) of internal bonding and MOR (*n* = five specimens from each board). EN 319 is the European standard for IB (0.35 N/mm^2) and the MOR (11.5 N/mm^2). Different letters indicate a significant different between mean values ($p < 0.0001$)

Several reasons make the storage stability of a binder system a critically important parameter in the wood panel industry. A binder with good storage stability allows an adequate storage time on the production site while ensuring that panels maintain produced at different time intervals maintain the same mechanical properties. Also, a stable binder helps in maintaining structural integrity, preventing premature failure or early degradation of the panels. Secondly, binders with poor storage stability can lead to variability in the quality of the wood panels produced. This inconsistency can result in panels that do not meet industry standards, leading to product recalls or customer dissatisfaction. A stable binder will as well allow for more predictable performance, reducing the need for frequent adjustments in the production process. Thirdly, panels with compromised bonding strength due to binder instability may need to be discarded, leading to material wastage and increased production costs, leading to economical inefficiency. Using a stable binder minimizes the risk of producing substandard panels, thus reducing the costs production associated.

The nitrite-treated CPI-N-60 binder demonstrated significantly better storage stability compared to the CPI-B-0 binder (Fig. 4). Across all storage times, the CPI-N-60 variants consistently outperformed CPI-B-0, demonstrating higher IB values. This indicates that the nitrite treatment enhanced the internal bonding strength of the CPI-N-60-bonded particleboards.

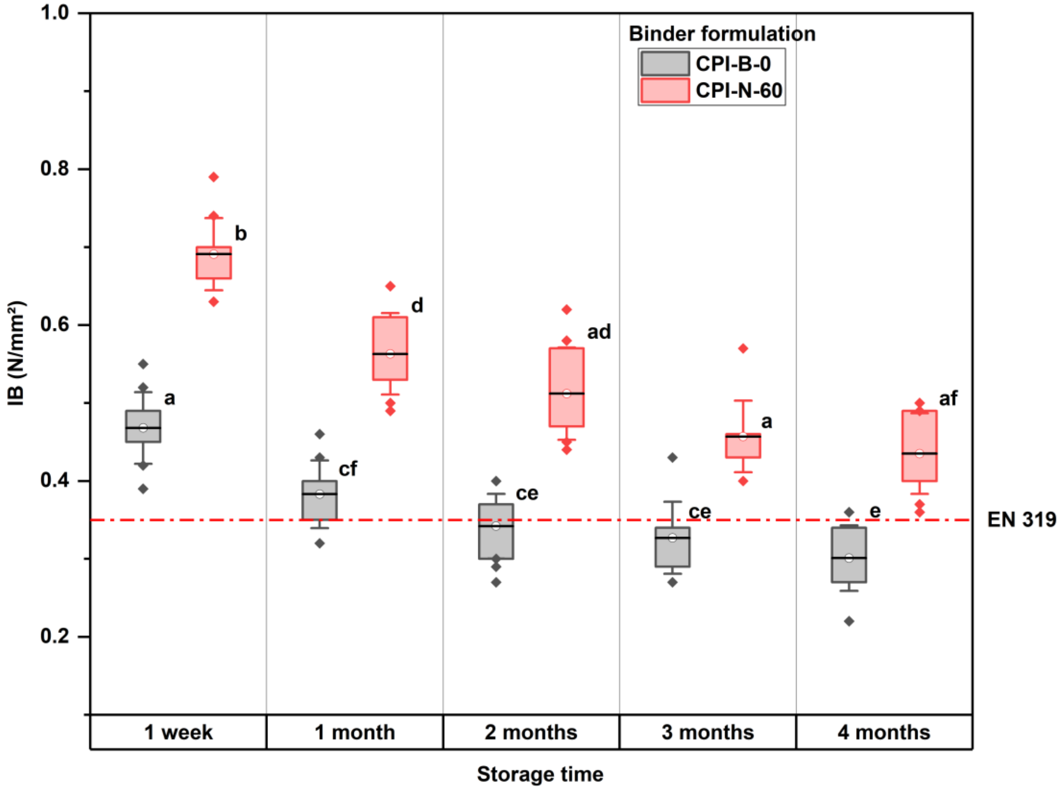

Fig. 4. Effect of binder storage time on the internal bonding strength of the one-layer particleboards. Box plots (25th quartile, mean and 75th quartile) and whiskers (1 × standard difference) of IB (*n* = five specimens from each board, thus 100 test specimens in total). EN 319 is the European standard for IB (0.35 N/mm^2). Different letters indicate a significant different between mean values ($p < 0.001$)

Over a storage period of up to four months, CPI-N-60 maintained higher internal bonding (IB) values, indicating superior long-term performance. This stability ensures that wood panels bonded with CPI-N-60 would perform reliably over extended periods, making it a preferable choice for manufacturing high-quality, durable wood panels. The storage time significantly affected the performance of both CPI-based binder formulations. After four months, the IB was reduced by about 37% for both variants, with values dropping from 0.69 to 0.43 N/mm^2 and from 0.47 to 0.30 N/mm^2 for the CPI-N-60 and CPI-B-0, respectively. The decrease rate was found to be higher within the first month for both binder variants and gradually slowed down as the storage time increased. The alkaline denaturation process of the protein can be reversible. The reduced performance of the adhesive over time is likely attributable to the oxidation process occurring during storage. By reducing the pH of the slurry, it allows the protein to refold at a certain degree, reducing the available functional groups in the binder. Despite this, with an IB value of 0.43 N/mm^2 after four months of storage, the CPI-N-60 was found to be a competitive candidate for replacing the conventional binding systems.

Unlike observed with the IB, the MOR and MOE performance of both CPI-based binder variants was comparable at every testing time (Fig. 5), the binder formulation having no significant effect. Both binders showed a decline in MOR over time, indicating that the bonding properties degraded with storage. However, CPI-N-60 generally maintained higher MOR values over extended storage periods compared to CPI-B-0. The recorded MOR values for both binder variants show variability over time, ranging from 7.4 to 11.4 N/mm² and from 8.3 to 12.4 N/mm² for CPI-B-0 and CPI-N-60, respectively. Although the observed trend was similar for both binders, the CPI-B-0 underwent higher oxidation/degradation. After four months, this adhesive variant had lost 20% of its performance, while the CPI-N-60 registered only a 15% decrease.

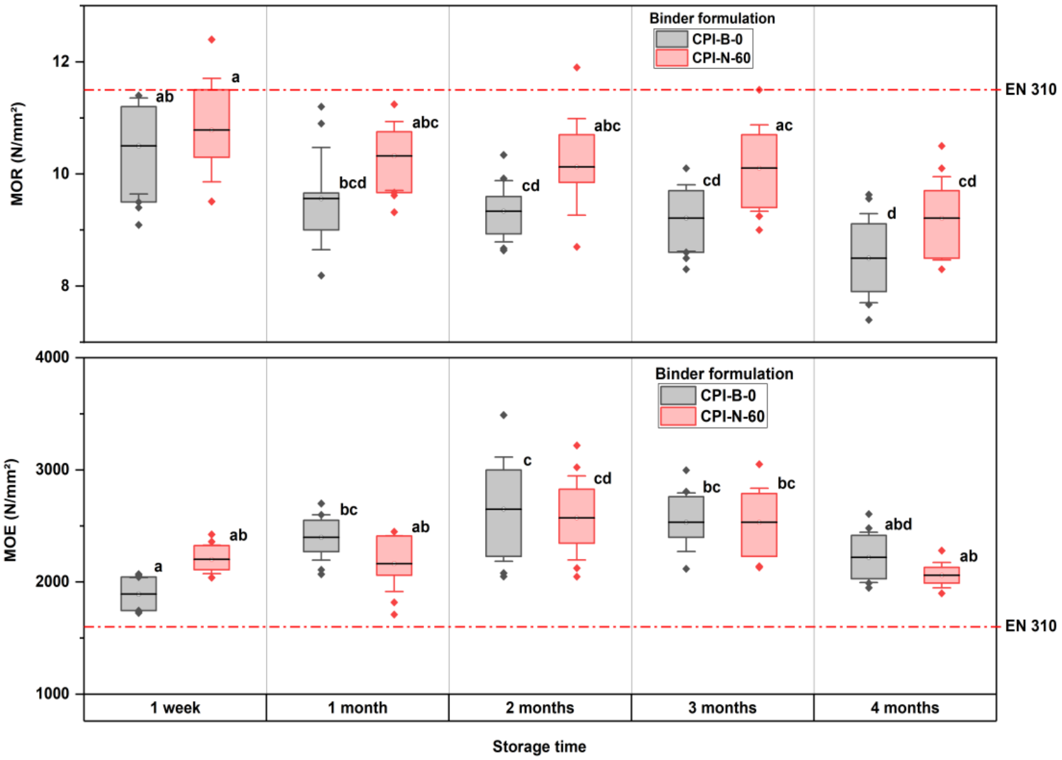

Fig. 5. Effect of binder storage time on the MOR and MOE of the on-layer particleboards. Box plots (25th quartile, mean and 75th quartile) and whiskers (1 × standard difference) of MOR (*n* = five specimens from each board, thus 100 test specimens in total). EN 310 is the European standard

for MOR (11.5 N/mm^2) and MOE (1600 N/mm^2). Different letters indicate a significant different between mean values ($p < 0.001$)

The MOE, or modulus of elasticity, is an important measure of the stiffness of the particleboard. It indicates the material's ability to resist deformation under load. The binder variant and storage time significantly impacted the MOE of one-layer particleboards. CPI-N-60 generally provided higher initial stiffness and better long-term stability compared to CPI-B-0, which showed a more pronounced increase in stiffness over the first two months but declined slightly afterwards. The MOE values for CPI-B-0 ranged from 1725 to 3490 N/mm^2 across all storage times, while that of the CPI-N-60 ranged from 1710 to 3218 N/mm^2. Initially, CPI-B-0 demonstrated lower MOE values at the 1-week mark. These values tended to increase with longer storage times, reaching a peak at 2 months. This suggests that the CPI-B-0 binder might have been undergoing some initial curing or cross-linking that strengthens the particleboard over time. However, after the peak, there was a slight decline observed at the 4-month mark, indicating possible degradation or over-curing that could compromise the material's stiffness. For CPI-N-60, the MOE values were generally higher initially and showed a more consistent performance over time. The values peaked at the 2-month mark as well but did not show as significant an increase as CPI-B-0. This suggests that CPI-N-60 had a more stable performance and might possess better initial curing characteristics and overall stability.

The same trend was observed with the mechanical properties of the CM-bonded particleboards, with values decreasing over time. The results indicate that CM-N-60 was more effective than CM-B-0 in terms of initial bonding strength (0.39 and 0.32 N/mm^2 respectively) and stability over time. However, both canola meal-based binders exhibited degradation over prolonged storage, with CM-B-0 showing a more pronounced decline (Fig. 6). Both binders showed higher IB values initially (at 1 week), with CM-N-60 outperforming CM-B-0, indicating superior immediate adhesive properties. CM-N-60 maintained relatively higher and more stable IB values over the first three months compared to CM-B-0, which showed a more noticeable decline in IB values after the second month, continuing into the fourth month. Both binders exhibited a decrease in IB values over time. However, CM-B-0 showed a more significant drop, particularly at the 4-month mark (with 56% decrease compared to 36% for CM-N-60), suggesting greater susceptibility to degradation over extended storage periods. The obtained values were lower compared to that of the CPI-bonded variants. This was however expected, as canola meal contains lower protein content (about 36% as reported by Ostendorf *et al.* 2021) and higher levels of non-protein components such as fiber, carbohydrates, and lipids compared to CPI. The presence of these non-protein substances can interfere with the adhesive properties of the protein, leading to weaker bonding. Moreover, the CPI is a more refined product with higher protein purity (90%), which enhances its reactivity and bonding capabilities. In contrast, canola meal contains various impurities that may reduce the overall reactivity and effectiveness of the adhesive. Also, the molecular structure of the proteins in canola meal may differ from that in CPI, affecting their ability to form strong cross-links. The denaturation and modification processes used to extract CPI often enhance its adhesive properties, which may not be as effective in the less processed canola meal. Canola meal-based adhesives may exhibit different aging behaviours compared to CPI-based adhesives. The degradation of proteins and other components over time can negatively impact the adhesive's performance, leading to lower IB values after prolonged storage.

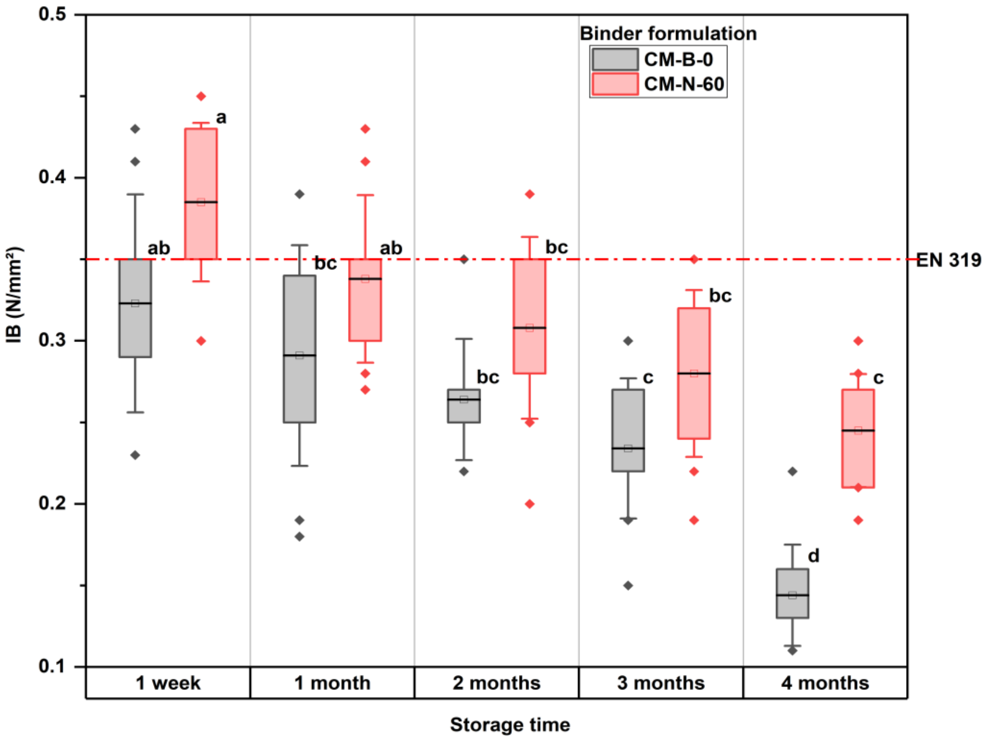

Fig. 6. Effect of binder storage time on the internal bonding strength of the one-layer CM-bonded particleboards. Box plots (25th quartile, mean and 75th quartile) and whiskers (1 × standard difference) of IB (*n* = five specimens from each board, thus 100 test specimens in total). EN 319 is the European standard for IB (0.35 N/mm^2). Different letters indicate a significant different between mean values (p < 0.001)

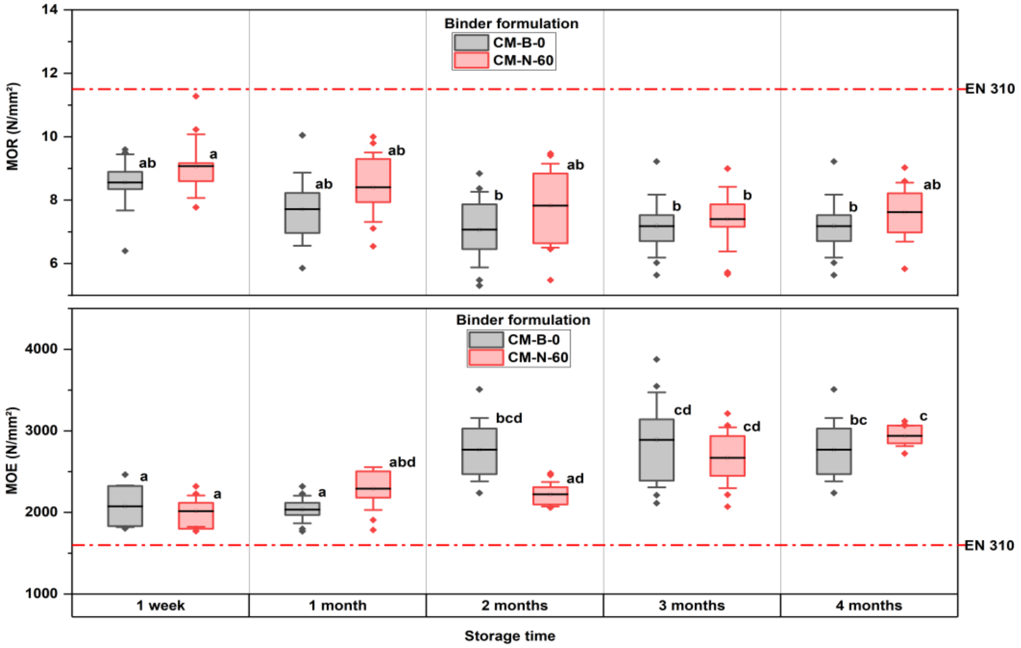

Fig. 7. Effect of adhesive storage on the MOR and MOE of the on-layer CM-bonded particleboards. Box plots (25th quartile, mean and 75th quartile) and whiskers (1 × standard difference) of MOR (*n* = five specimens from each board, thus 100 test specimens in total). EN 310 is the European standard for MOR (11.5 N/mm^2) and MOE (1600 N/mm^2). Different letters indicate a significant different between mean values (*p < 0.001*)

Both canola meal-based binders exhibited a decrease in MOR values over prolonged storage, with CM-B-0 showing more significant degradation (Fig. 7). The CM-N-60 variant demonstrated a better performance than CM-B-0, particularly in terms of initial MOR values and stability. Both binders also showed degradation over prolonged storage, with CM-B-0 being more susceptible. Comparatively, CPI-based binders provided higher and more stable MOR values, indicating their superior performance for long-term applications. These insights are crucial for optimizing binder selection in the wood panel industry to enhance product quality and durability. The MOE of the CM-bonded particleboard variants was comparable to that of the CPI-bonded, however with lower values recorded. CM-N-60 demonstrated higher initial MOE values and better stability over time compared to CM-B-0. Both canola meal-based binders exhibited fluctuations in MOE over prolonged storage.

CONCLUSION

This study investigated the effects of different canola-based binder formulations and their storage times on the mechanical properties of one-layer particleboards, focusing on internal bond strength (IB), modulus of rupture (MOR), and modulus of elasticity (MOE). The binder formulations examined were canola meal-based (CM-B-0 and CM-N-60) and canola protein isolate-based (CPI-B-0 and CPI-N-60).

1. The findings revealed that canola protein isolate-based binders (CPI-B-0 and CPI-N-60) consistently outperformed their meal-based counterparts (CM-B-0 and CM-N-60) across all measured properties (IB, MOR, and MOE). This suggests that CPI binders offer superior performance and stability, making them more suitable for applications requiring high strength and durability.
2. While both binder types exhibited some degree of degradation in mechanical properties over time, CPI binders maintained higher values and experienced less significant fluctuations compared to CM binders. This indicates better long-term performance and resistance to biodegradation.
3. Among the canola meal-based binders, CM-N-60 generally performed better than CM-B-0, suggesting that the specific formulation provides enhanced adhesive properties and stability. The canola meal-based binders, CM-B-0 and CM-N-60, showed varying IB values over time. CM-N-60 generally exhibited higher and more stable IB values compared to CM-B-0, suggesting better adhesive properties and durability. In contrast, the canola protein isolate-based binders consistently showed higher IB values compared to the canola meal-based binders, indicating superior bonding capabilities and stability over extended storage periods.
4. For MOR, both CM-B-0 and CM-N-60 displayed fluctuations over time, with CM-N-60 maintaining relatively higher and more stable values than CM-B-0. However, significant variations were observed in both binder types as storage time increased. The CPI binders outperformed CM binders in terms of MOR, maintaining higher values throughout the storage period. This indicates better structural integrity and load-bearing capacity.

5. Regarding the MOE, CM-N-60 exhibited higher initial values and greater stability over time compared to CM-B-0. Nonetheless, both binders showed fluctuations in MOE, with CM-B-0 experiencing more significant variations, especially after extended storage. The CPI binders consistently provided higher MOE values compared to CM binders, demonstrating better stiffness and elasticity over time.

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Author Contributions

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Conflict of Interest

The authors declare no conflict of interest. The funders had no role in the design of the study; in the collection, analyses or interpretation of data; in the writing of the manuscript; or in the decision to publish the results.

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