https://doi.org/10.26525/jtfs2025.37.4.427
ISSN: 0128-1283, eISSN: 2521-9847

PHYSICAL AND MECHANICAL PROPERTIES OF *SALACCA* FROND PARTICLEBOARD BONDED WITH EXPIRED CITRIC ACID BINDER

GK Dewi*, R Dwiyanna, VA Qibtiya & R Widyorini

Department of Forest Product Technology, Faculty of Forestry, Universitas Gadjah Mada, Bulaksumur, Yogyakarta, Indonesia 55281

**greitta.k.d@ugm.ac.id*

Submitted February 2025; accepted March 2025

The performance of wood binders in particleboard is diminished by both expiration and improper storage. Those effects on citric acid powder, which is one of potential natural adhesives of particleboard for the future, is still unknown. This research investigated the effect of duration length of citric acid past the expiration and its storage conditions on the properties of *Salacca* frond particleboard. The effect of its binder content and pressing temperature was also evaluated. One and two-years expired citric acid were used, especially for two-years expired citric acid, two types storage conditions were conducted, namely unexposed to ambient humidity (close-lid container storage) and exposed to ambient humidity (open-lid container storage). The results showed that the water resistance of the particleboard decreased the longer citric acid passed its expiration and aggravated with exposed circumstances (open-lid). Under sufficient binder content and optimal pressing temperature, the mechanical properties of *Salacca* frond particleboard bonded with expired citric acid were retained in the properties requirement by Japanese Industrial Standard (JIS) A 5908. Expired citric acid still has the potential to be used as a natural binder for particleboard until 2 years passed the expiration.

Keywords: Expired citric acid, duration length, storage condition, binder content, particleboard.

INTRODUCTION

In the current increasing awareness on Sustainable Development Goals (SDG's) and environmental issues through the Paris Agreement, the use of environmentally friendly material and waste management become highly appreciated globally. Consequently, lignocellulose-based materials such as wood for structural and non-structural applications have new highlight due to their renewability and eco-friendly properties. This reflected in the rising demand and production of wood-based panels, particularly particleboard. According to BPS-Statistics Indonesia, particleboard production in Indonesia increased by 231% from 61,249.9 m³ in 2019 to 202,644.92 m³ in 2023 (BPS-Statistic Indonesia 2020, 2024). Similarly, Food and Agriculture Organization (FAO) reported a global increase in particleboard production from 130,913,798 m³ in 2019 to 160,598,927 m³ in 2023 (FAO 2019, 2023).

Particleboard was mainly manufactured from adhesive and various types of lignocellulose particles, mostly from wood residues, but nowadays it is being developed using agricultural crop waste as homogenous and/or heterogeneous particle composition, for example sengon (*Paraserianthes falcataria*) wood, bagasse, and their mixtures; spruce wood; oil palm biomass and *Salacca* frond waste (Dewi et al. 2022, Idris et al. 2024, Laksono et al. 2022, Pędzik et al. 2021, Widyorini et al. 2018). *Salacca* frond was used as a raw material, and the effects of citric acid-maltodextrin composition ratio, pressing temperature, and pressing method were investigated (Widyorini et al. 2018). The boards were manufactured under the conditions as follows: single-step and three-step press methods, adhesive content of 20 wt%, and pressing temperature of 180 and 200 °C. *Salacca* frond waste was known for its high lignocellulose content, its potency in the quantity and periodic availability, and its high particleboard quality (Dewi et al. 2022, Widyorini et al. 2018). The utilisation of sustainably managed waste can serve as an alternative substitute in the production of renewable bio-based particleboard (Oh 2020).

More than 90% of particleboards use formaldehyde-based adhesive (Flores et al. 2011), in spite of a long history of natural adhesive development. For example, soybean protein adhesive that had been developed for plywood in the USA since 1930 (Zhang et al. 2023). It was due to the advantages of formaldehyde-based adhesive, namely its superior bonding performance, efficient procedure, and feasibility (Hussin et al. 2022). Therefore, discovery of natural adhesives or binder with comparable performance to formaldehyde-based adhesives are still on going. Especially on particleboard, some natural adhesives were reported to be potentially used such as lignin, tannin, protein-based (casein or soy flour), carbohydrate-based, natural rubber latex, vegetable oil, and citric acid-based adhesives (Umemura et al. 2017, Huaxu et al. 2021, Islam et al. 2022, Hussin et al. 2022, Patel et al. 2024). Moreover, Pizzi et al. (2020) classified citric acid as a promising natural adhesive due to its superior water resistance and strength, as well as being compatible with many raw materials for particleboard, such as wood (recycled wood, softwood sawmilling residues), and non-wood (bamboo, rubberwood, *Salacca* frond) (Umemura et al. 2012, Huaxu et al. 2021, Widyorini et al. 2016, 2018, Scharf et al. 2024). Meanwhile, low cost of manufacture, consistent upward trend of global production, ease of availability, non-toxicity, biocompatibility, universality, and the safety of its breakdown products are what define citric acid (Apelblat 2014, Książek 2024). To improve the performance of citric acid as wood binder, citric acid had been developing with a mixture of some additives, such as sucrose (Wibowo et al. 2021), starch (Huang et al. 2022), maltodextrin (Widyorini et al. 2018), tannin (Li et al. 2023), etc.

The effectiveness of adhesive performance was influenced by the application procedure towards the glue catalog recommendations, storage stability (Liu et al. 2018), and is in the use by date or shelf life. The application procedure comprises the optimal adhesive mixture, adhesive content, adhesive spreading methods and press methods (pressing temperature, pressing time, and pressure). Moreover, long-term storage stability was required in adhesive. It was related to period time and the storage environment, such as specified temperature, relative humidity, visible light, ultraviolet radiation, and container closures/seal (open/closed) (Simmons 1995). The container seals are important to provide protection against fluid's damage and contamination by minimising dehydration and rehydration, as well as protecting fluid from light, ultraviolet, and impurities (Simmons 1995). Dehydration and rehydration related to moisture promote the instability of organic solids chemically and physically (Kwok et al. 2010). Arendse and Jideani (2022) also state that storage environment, water activity, temperature, and the environment along with the ingredients and the production process affect the shelf life of a material.

Discrepancy with the application procedure recommendations and optimal storage environment and also expired conditions could decrease the performance of adhesive. The decrease could be through chemical change (structure change) and physical change that affect the bonding process of adhesive. This could be severe if the adhesive had already been in the mixture solution in which the solvent, hardener, filler or other additives have been added. Urea formaldehyde (UF) was known to have structure change and increase its viscosity during storage at room temperature and worsen after past the expiry time, therefore affecting the processability and performance of the adhesive (Simer et al. 2005, Aini et al. 2024, Kim et al. 2001). The viscosity of natural rubber and cottonseed meal-based adhesives (CSM) also increases significantly in a longer storage time (Radabutra et al. 2020, He et al. 2016). This increase in viscosity decreased the lap shear strength remarkably in the wood-lamination with natural rubber adhesive, but only slightly decreased the lap shear strength with CSM adhesive. The optimal manufacturing conditions might also change caused by the chemical changes and the physical changes of the adhesives.

Performance of expired citric acid binder and its storage condition on the particleboard had not been investigated yet. In addition, citric acid

Table 1 Moisture content of powder citric acids and pH of its solutions

Binder condition	Moisture content (%) of powder citric acids	pH of citric acid solutions
Unexpired citric acid solution	0.28±0.03	0.62±0.01
2-years passed the expiration (closed-lid)	0.54±0.10	0.60±0.02
2-years passed the expiration (open-lid)	3.58±0.35	0.63±0.01

binder was stored in crystalline powder form, different from the commercial formaldehyde-based adhesives which had been generally mixed with solvent. Moreover, anhydrous citric acid was known to absorbs moisture in a specific relative humidity (Książek 2024). Our study aims to evaluate the performance of the expired citric acid at some conditions (1 year and 2 years passed the expiration in closed-lid, and 2-years passed the expiration in open-lid storage) on the properties of *Salacca* frond particleboard. Comparison of those properties with the properties of the effective citric acid particleboard (used by date, unexpired citric acid) in the same raw material studied by Widyorini et al. (2018) was also conducted.

MATERIALS AND METHODS

Salacca frond waste from the pruning activities of *Salacca* plantation was acquired from Turi, Cangkringan, Sleman and Yogyakarta. The frond was then chipped, air-dried until the moisture content of ±12%, crushed in a ring knife flaker, and sieved on a 10-mesh sieve for the raw material preparation of particleboard. Citric acid (Brataco LLC, Banten, Tangerang, Indonesia) that expired for one year and two years past its expiration (kept in a closed-lid container and stored in a dry cabinet at 35% relative humidity, 28°C temperature) were used as a binder. The 2 years-expired citric acid that is exposed to room relative humidity of ±71% (open-lid container storage) was also used for representing the exposed citric acid during the storage toward the environment.

The preparation of binder solution followed the citric acid solution preparation developed by Umemura et al. (2012). The citric acid at three conditions (1-year passed the expiration closed-lid, 2-year passed the expiration-closed lid, and 2-year passed the expiration-open lid) were dissolved in the distilled water at 60 wt% concentration. The pH of each condition was measured using a Mettler Toledo Seven Compact S220 pH Meter, as presented in Table 1. The binder solution prepared at 10 and 20 wt% based on the air-dried particles weight were evenly mixed with *Salacca* frond particles. Furthermore, the binder-coated particles were oven-dried at 80°C for 4 hours until its moisture content was around 6%. Subsequently, it was arranged into a cube-shape mat with the 25 cm × 25 cm size. The mat was then hot-pressed using 3-step press methods developed by Widyorini et al. (2018) at 180 and 200°C pressing temperature for 10 minutes with a specific pressure of 3 MPa. The final particle board had the target size of 25 cm × 25 cm × 1 cm and target density of 0.8 g/cm^3. The particleboard manufacture treatment in this study was concluded in Table 2. Each treatment was conducted in triplicate, therefore the total particleboard made was 36 boards.

The particleboard was conditioned for 1 week in a room with an ambient temperature of 27°C and relative humidity of 71%. Each board was then sawn using a circular saw into eight samples, as illustrated in Figure 1. Sampels of 5 cm × 5 cm size (No. 1–4 and 6–8) were used for internal bonding strength, water absorption, moisture content, and density tests, while a 20 cm × 5 cm size sample (No. 5 and 10) were used for bending tests. Each test sample is chosen whose density represents the board's average density. Density, moisture content (MC), bending strength and internal bonding strength (IBS) were tested according to Japanese Industrial Standard (JIS) A 5908 (2015), but be specifically modified in the sample size of moisture content and density. Furthermore, a water absorption test at 24 hours water immersion was conducted according to Widyorini et al. (2018).

All data of the board properties were statistically analysed to determine the effect of the binder conditions, binder content and pressing temperature of particleboard

Table 2 The treatment arrangement

Binder content (wt%)	Pressing temperature (°C)	Binder condition	Treatment*
10	180	1-year passed the expiration (closed-lid)	10-180-1C
		2-years passed the expiration (closed-lid)	10-180-2C
		2-years passed the expiration (open-lid)	10-180-2O
	200	1-year passed the expiration (closed-lid)	10-200-1C
		2-years passed the expiration (closed-lid)	10-200-2C
		2-years passed the expiration (open-lid)	10-200-2O
20	180	1-year passed the expiration (closed-lid)	20-180-1C
		2-years passed the expiration (closed-lid)	20-180-2C
		2-years passed the expiration (open-lid)	20-180-2O
	200	1-year passed the expiration (closed-lid)	20-200-1C
		2-years passed the expiration (closed-lid)	20-200-2C
		2-years passed the expiration (open-lid)	20-200-2O

*10 and 20 = 10% and 20% binder content; 180 and 200 = 180°C and 200°C pressing temperature; 1C = citric acid binder with 1-year passed the expiration and closed-lid storage; 2C = citric acid binder with 2-year passed the expiration and closed-lid storage; 2O = citric acid binder with 2-year passed the expiration and open-lid storage. E.g., 10-180-1C = 10% binder content, 180°C pressing temperature, citric acid binder with 1-year passed the expiration and closed-lid storage

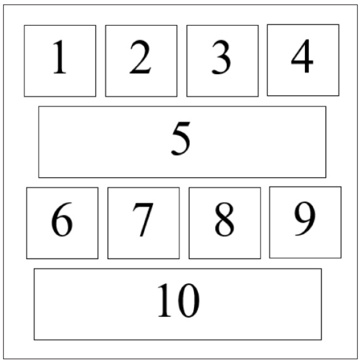

Figure 1 Sample sawing patterns

manufacture on the properties of expired citric acid bonded particleboard. Analysis of variance was conducted and then continued with Tukey's honestly significant difference test at 1% and 5% test levels. Relationships between particleboard properties and groups within the particleboards were evaluated using Principal component analysis (PCA). Kaiser-Meyer-Olkin (KMO) Measure of Sampling Adequacy and Bartlett's Test of Sphericity was conducted before PCA to verify the suitability of the dataset for factor analysis. The KMO values was over 0.5 (0.584), and the Bartlett's test significance was <0.001 (2.01×10^{-7}), therefore PCA was suitable to be conducted.

RESULTS

Salacca frond particleboards were successfully made using expired citric acid binder. The density of the expired citric acid-bonded particleboard ranged from 0.64–0.78 g/cm^3. The effect of binder content, pressing temperature, binder condition, and their combinations on the properties of particleboard were shown in Table 3. Analysis of variance showed that the interaction between binder content, pressing temperature and binder condition significantly affected moisture content (MC), internal bonding strength (IBS), and modulus of elasticity (MOE) of the particleboard ($p<0.05$).

Table 3 The ANOVA results

	The Significance value				
	Moisture content	Water absorption	Internal bonding strength	Modulus of rupture	Modulus of elasticity
Binder content	3.8×10^{-2}*	2.5×10^{-5}**	8.9×10^{-7}**	1.1×10^{-4}*	9.7×10^{-6}**
Pressing temperature	1.2×10^{-1}	1.1×10^{-7}**	5.7×10^{-2}	8.0×10^{-1}	2.4×10^{-4}**
Binder condition	6.6×10^{-16}**	9.4×10^{-13}**	9.4×10^{-5}**	8.5×10^{-1}	1.1×10^{-6}**
Binder content*Pressing temperature	1.4×10^{-1}	1.3×10^{-1}	2.0×10^{-2}*	6.4×10^{-1}	3.5×10^{-5}**
Binder content*Binder condition	6.4×10^{-1}	7.7×10^{-3}**	1.7×10^{-3}**	1.5×10^{-1}	3.1×10^{-2}*
Pressing temperature*Binder condition	3.8×10^{-5}**	1.2×10^{-2}*	2.8×10^{-3}**	6.9×10^{-1}	1.7×10^{-8}**
Binder content*Pressing temperature*Binder condition	3.2×10^{-2}*	5.5×10^{-1}	1.3×10^{-2}*	6.0×10^{-1}	1.2×10^{-2}*

* = significant at 5% test level and ** = significant at 1% test level

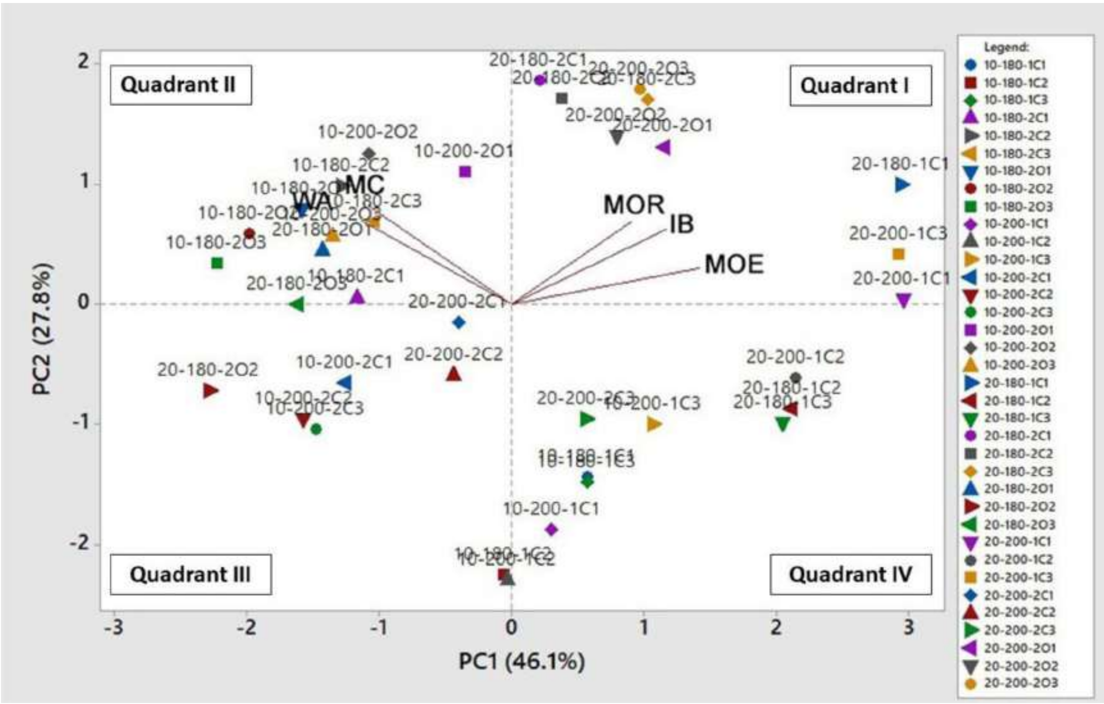

Figure 2 Biplot score PCA of particleboard bonded with expired citric acid

Principal component analysis (PCA) presented two principal components (PC1 and PC2) explained 73.9% of the total variance in the dataset (Figure 2). Similar to Mehrvan et al. (2024), PC1 showed positive correlations with mechanical properties (such as modulus of rupture, modulus elasticity and internal bonding strength), and a negative correlation with water absorption. Our study added that moisture content was parallel to water absorption and has negative correlation with mechanical properties. This showed that particleboards with high mechanical properties tend to have high resistance of moisture and water. Figure 2 also describes that particleboards in quadrant I relatively have high mechanical properties, while particleboards in quadrant II have relatively high-water absorption and moisture content.

Moisture content (MC) and water absorption

(WA) of *Salacca* frond particleboards with 1 year-expired citric acid tend to have low values both in two temperatures and two binder contents (Figure 3 & 4). Then, the longer the citric acid expires, the higher the MC and WA. High IBS, MOR, and MOE values were also achieved at 1 year-expired citric acid at 20 wt% binder content (Figure 5–7). The longer the citric acid expires, the decrease the mechanical strength values tend to be.

DISCUSSION

Expired citric acid binder is still effective as a natural binder for particleboard, although the particleboard manufactured has lower density than the target density. Lower density than the target density set in this study was expected mainly due to board widening at non-moulding particleboard manufacturing. This could have occurred because of the low adhesion of binder

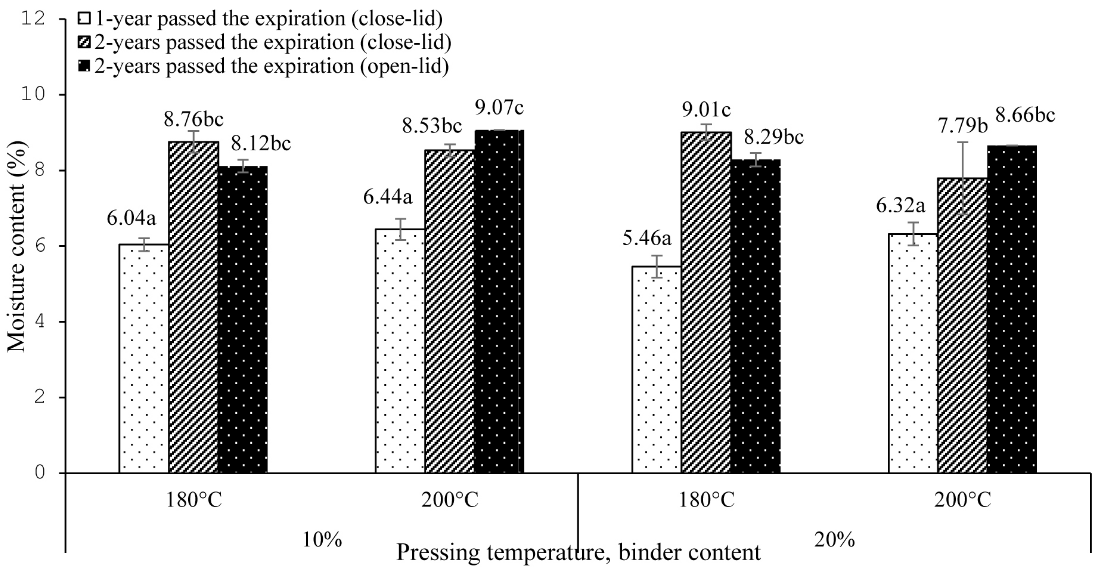

Figure 3 Moisture content of expired citric acid-bonded particleboard made from *Salacca* frond

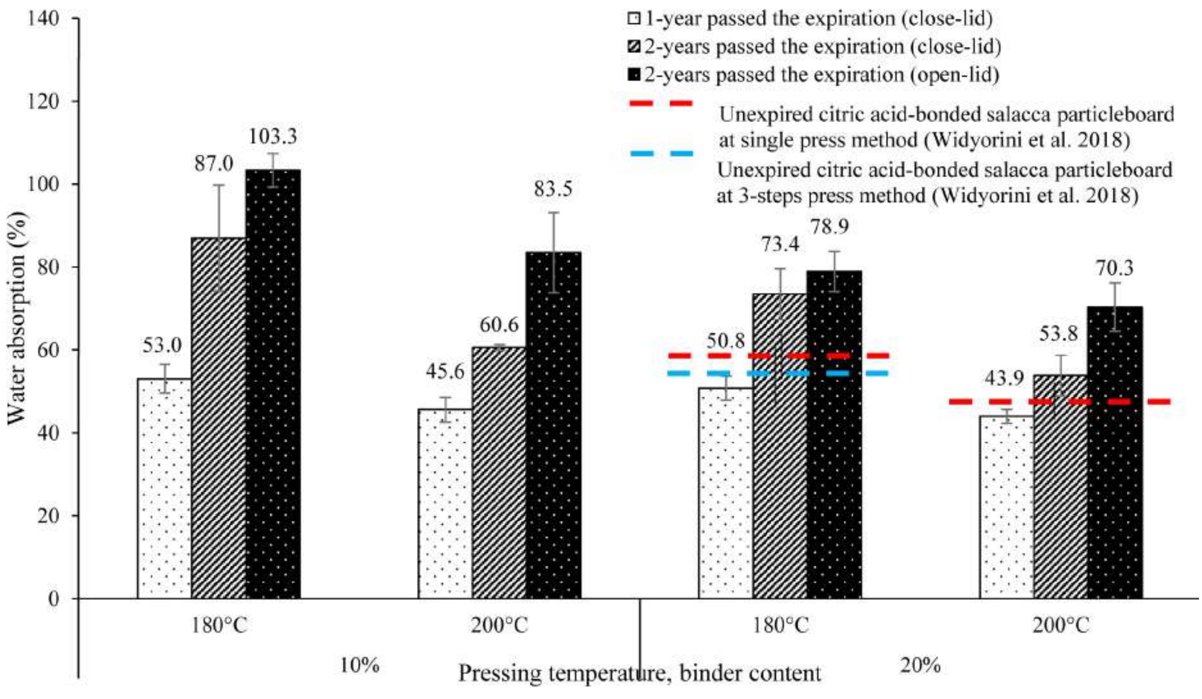

Figure 4 Water absorption of expired citric acid-bonded particleboard made from *Salacca* frond

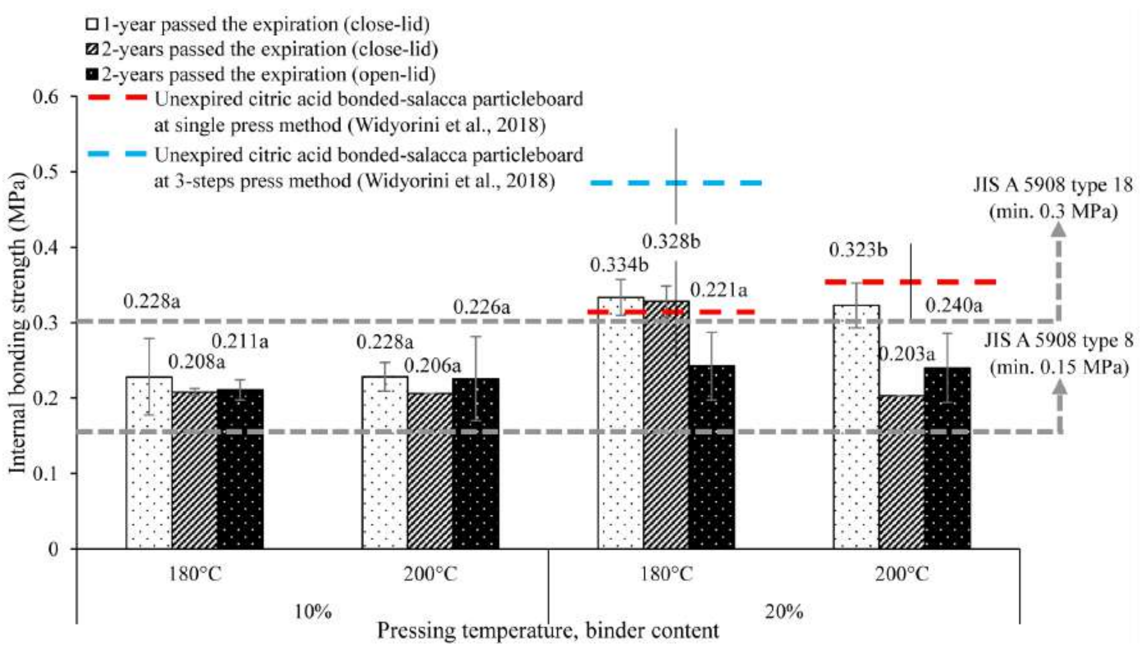

Figure 5 Internal bonding strength of expired citric acid-bonded particleboard made from *Salacca* frond

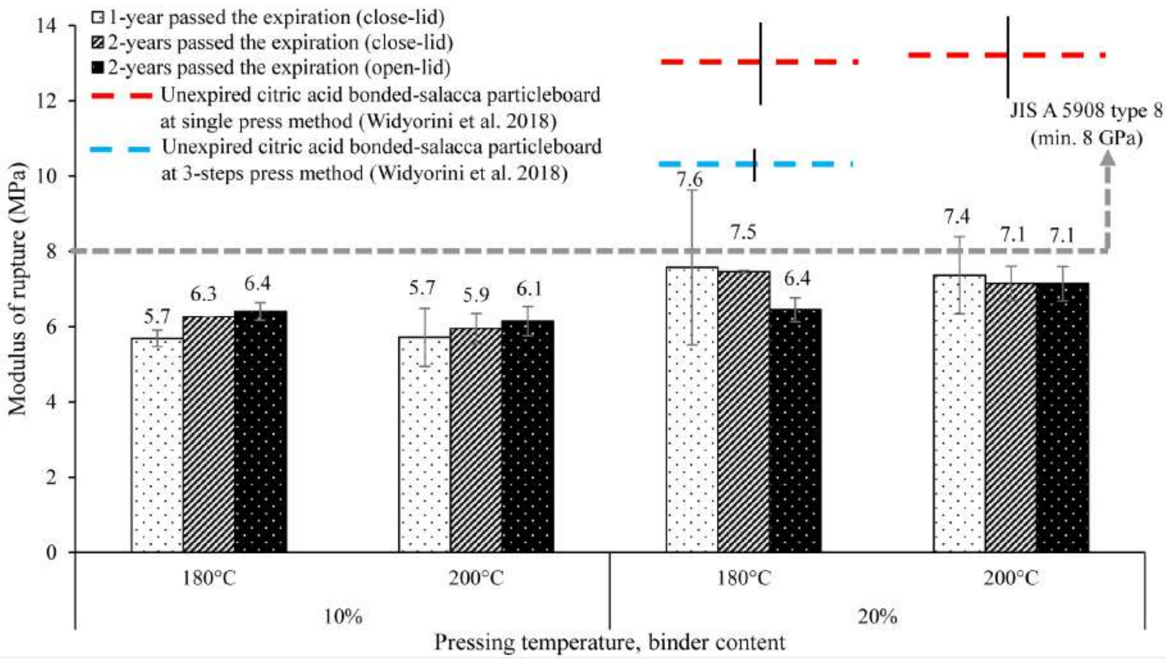

Figure 6 Modulus of rupture of expired citric acid-bonded particleboard made from *Salacca* frond

before the hot press, therefore low bonding among particles was obtained. Citric acid was known to actively bind the lignocellulose for particleboard manufacturing at a temperature of 180°C (Lee et al. 2020). Low tackiness of the expired citric acid binder might also play a role in a low particle bonding before hot-press. Board widening was also found in sucrose-citric acid-based particleboard made from sugar palm dreg (Aisyadea et al. 2023), and maltodextrin-based particleboard made from *Salacca* frond (Dewi et al. 2022). Some wood adhesive might be classified as latent adhesives that are ineffective at ambient temperature and acquire a specific activation to bind, namely high temperature, and hardener.

Table 3 showed all the board properties affected by binder content, linear with previous studies (Zeleniuc et al. 2019, Umemura et al. 2012). This showed that expired condition of citric acid did not change the effect of binder content on particleboard properties. The binder condition and the interaction between binder condition and pressing temperature affects significantly almost all the particleboards, except MOR. It was expected that binder conditions could make different characteristics and properties of binder, therefore affected the pressing temperature required to melt the binder and bind the particles. Our early study found that the moisture content of expired citric acid powder tends to be different among different binder conditions (Table 1). This might affect binder solid content and moisture content of the mat. Combination between material's moisture content and pressing temperature affect the IBS and TS of binderless particleboard made from poplar wood (Cheng et al. 2024).

Pressing temperature remarkably affected the WA and MOE of particleboard bonded with expired citric acid (Table 3). In particleboard manufacturing, pressing temperature affects the physical and chemical properties of both raw material and the binder. It might different between expired and unexpired binder. However, low pressing temperatures could make the adhesive uncured in high temperature-cured binder, such as citric acid, conversely higher pressing temperature than the required pressing temperature could cause over-cured adhesive, and both conditions lead to low strength of particleboard (Malanit et al. 2009). Hill (2006) added that the use of temperature within the range of 180–206°C can alter the chemical components of lignocellulose, especially the cellulose and hemicellulose decomposition and rearrangement. D'Amico et al. (2010) discovered that curing or pressing temperature firmly affects the adhesive properties of wheat flour polymers and the formation of wood-to-wood bonding (the interfacial bonding). The strength of interfacial bonding was related to MOE of particleboard. MOE is related to bonding energies within the material (Stoeckel et al. 2013, Tabor 1951). Related to interfacial bonding, the resistance ability of bonding towards the water immersion determines the WA values. Water might bind with the free hydroxyl groups of the wood particles, making the particle absorbing and swelling, promoting the interfacial failure between the binder and particle if the bonding strength was low, and then causing the WA of the particleboard. In addition, PCA showed a high correlation between MOE and WA in negative direction (Figure 2).

The moisture content (MC) ranged from 5.16 to 9.16% (Figure 3). This range MC was similar with the range of MC of other *Salacca* particleboard but using maltodextrin binder, namely 5.75–7.59% (Dewi et al. 2022) and other citric acid *Salacca* composite board but made from alkaline-modified *Salacca* fibrovascular, namely ±6% (Hakim et al. 2024). Additionally, those MC were within the range of commercial particleboard manufactured in Indonesia (8.35–10.85%), but lower than commercial particleboard manufactured in North America (10.9–12.0%) (Dettmer & Smith 2015, Wardani et al. 2015). Relative humidity, adhesive type, press temperature and particle size affected the MC of particleboard (Mo et al. 2001, Iskanderani 2008). All the MC in this study also met the JIS A 5908 standard that required the MC range of 5–13%. This showed that the expired citric acid up to 2-years passed the expiration, both in closed- or open-lid storage was still well-performed as a natural binder for particleboard as long as the sufficient binder content and the pressing temperature of 180 and 200°C were applied. However, the MC has increased remarkably from 1-year to 2-years past the expiration, and continues to increase when stored in open-lid storage. This was expected to affect the mechanical properties of particleboard, such as MOR, MOE, and IBS in inverse relationship (Halligain & Schniewiind 1974) and were proven in PCA results (Figure 2).

The WA of the expired citric acid-bonded particleboards (Figure 4) was 42.7–107.8%. Almost all WA values satisfied the acceptable WA value range of 20–75% (Anosike-Francis et al. 2022), except the WA from the treatment of the 180°C pressing temperature, 10% citric acid, and 2 years passed the expiration in both closed and open lid storage (87% and 103.3%), the WA from the treatment of the 200°C pressing temperature, 10% citric acid, and 2 years passed the expiration (open lid storage) binder condition (83.5%), as well as the WA from the treatment of the 180°C pressing temperature, 20% citric acid, and 2 years passed the expiration (open lid storage) binder condition (78.9%). Under the same pressing temperature, similar WA values were obtained between the 1 year expired citric acid and unexpired citric acid produced by Widyorini et al. (2018) at 20% citric acid (Figure 4). The WA value of unexpired citric acid-bonded particleboard was 55–58% and 46% at 180°C and 200°C pressing temperatures, respectively (Widyorini et al. 2018). Some commercial particleboards manufactured in North America have 44.3–47.7% WA values, but there have also been reports of WA values below 20% (Dettmer & Smith 2015). Expired citric acid still creates a good water-resistant bonding between particles in particleboard manufacturing, similar to unexpired citric acid.

WA values were remarkably increased from 1 year to 2 years after the expiration. However, more increase in WA values occurred at open-lid storage in 2-years past the expiration binder. This is in line with the moisture content trend. It is suspected that environmental factors, namely relative humidity, affect the moisture content of powder citric acid (Table 1), then affect physical and chemical stability of citric acid, therefore changing the reactivity and influencing the bonding among particles. Further research is needed to analyze the chemical changes that occur in citric acid after the expiration and their relations with water-related properties.

As the binder content increases, WA values decrease. This is comparable with the effect of increasing pressing temperature. The WA values decreased by 4.2–23.6% when citric acid content was increased from 10wt% to 20wt% at 180°C pressing temperature and by 10.2–13.6% at 200°C pressing temperature. However, the decrease rate of WA was 6.5% for the Salacca frond particleboard bonded with unexpired citric acid and pressed at 180°C (Widyorini et al. 2019). A significant decrease rate of WA was found in Huaxu et al. (2021) when the decrease rate was ∞ value (board disintegrated) to 137.93% at changing 10wt% to 20wt% in citric acid bonded particleboard made from rubber wood and pressed at 180°C.

All IBS values met the JIS A 5908 type 8 standard (min. 0.15 MPa), even some IBS values met type 18 (min 0.3 MPa). The IBS of the 1-year expired citric acid at 20% binder content was comparatively within the range of the unexpired citric acid's IBS values, which was 0.31 MPa and 0.35 MPa, as investigated by Widyorini et al. (2018) at single step hot press of 180°C and 200°C. That result was consistent with the WA properties. However, the IBS value of this study was 30% lower than that of the unexpired citric acid particleboard pressed using the 3-step method and 180°C by Widyorini et al. (2018) (±0.48 MPa). Hence, they still met the JIS A 5908 (2015) type 18 and resembled the IBS value of one commercial particleboard manufactured in North America (0.32 MPa) (Dettmer & Smith 2015). It showed that 1-year expired citric acid stored in a closed-lid container still has good performance as a wood binder. However, the longer the citric acid expires, the lower the IBS. The IBS values could decrease up to 37% from 1-year past the expiration to 2-year past the expiration in the condition of a closed-lid container. Similar trend occurred in particleboard with thick spent sulfite liquor-wheat flour where a slightly decreased IBS from fresh adhesive to the 30 days old adhesive (Ferreira et al. 2018). Additionally, the effect of open or closed-lid container at 2 years past expiration was different based on binder content and pressing temperature.

Even though the WA and IBS of the particleboard with the 1-year expired citric acid was similar with the unexpired citric acid-bonded particleboard at single press method, all modulus of rupture (MOR) and modulus of elasticity (MOE) of the expired citric acid-bonded particle board were remarkable lower than those of the unexpired citric acid particleboard at both single press and 3-steps press methods (Figure 6–7). The MOR of salacca particleboard bonded with unexpired citric acid was around 13 MPa at 180°C and 200°C single hot presses, meanwhile around 10 MPa at 180°C three step hot presses. Moreover, the MOE of *Salacca* particleboard bonded with unexpired citric acid was around 0.36 GPa at 180°C and 200°C single hot presses and around 2.8 GPa at 180°C three step hot presses (Widyorini et al. 2018). This might be caused by different particle geometry between 2 studies though both researches used the same passed through 10 mesh particle size. However, the particle geometry comparison is not possible due to the absence of the comparative data. Particle geometry with high slenderness ratio,

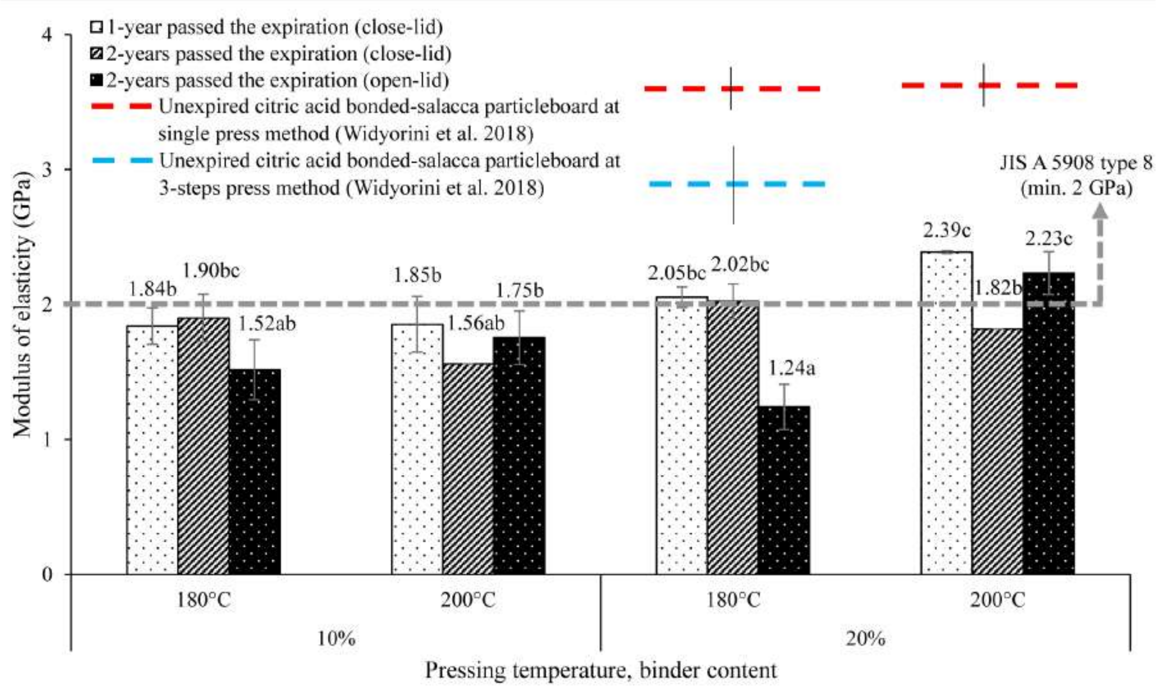

Figure 7 Modulus of elasticity of expired citric acid-bonded particleboard made from *Salacca* frond

a ratio between length and thickness, showed a high MOR and MOE strength of particleboard. In order to meet MOR and MOE standard, a poplar particleboard with 0.69 g/cm^3 and 8.09% adhesive content had to use particles with slenderness ratio of 47 (Arabi et al. 2023).

All MOR failed to satisfy the requirement standard of JIS A 5908 (2015) (8 MPa for type 8). Nevertheless, four MOEs satisfied JIS A 5908 type 8 (min. 2 GPa). These were 20% citric acid content after one year of expiration at both pressing temperatures (2.05 and 2.02 GPa), 20% citric acid after two years of expiration (close-lid) at 180°C pressing temperature (2.39 GPa), and 20% citric acid after two years of expiration (open-lid) at 200°C pressing temperature (2.23 GPa). This phenomenon also happened in particleboard manufactured from a mixture of sawdust and bamboo leaves bonded with urea formaldehyde, as studied by Olawale et al. (2024). Some of these particleboards have a modulus of elasticity of more than 2 GPa, although the modulus of rupture has not yet reached 8 MPa. Some improvement might be conducted to increase this particleboard's bending strength, especially the MOR, namely increasing the particleboard density or face layer density (Divkolaei et al. 2024, Korai 2022), increasing the slenderness ratio of the particles (Arabi et al. 2023), etc.

CONCLUSION

Expired citric acid still have potential to be utilized as a natural binder for particleboard. Particleboard bonded with citric acid after one year expiration exhibits internal bonding strength and water absorption that are still within the same range as those of unexpired citric acid-bonded particleboard at treatments of 20% citric acid, single press method and two pressing temperatures (180 and 200°C). Those particleboard properties satisfied JIS A 5908 standard type 8 and acceptable water absorption value, except the modulus of rupture. Interaction between binder condition, binder content, and pressing temperature affected the moisture content, internal bonding strength and MOE of the expired citric acid-bonded particleboard. Water resistance properties and mechanical properties of *Salacca* frond particleboard tend to decrease with the longer the citric acid passes its expiration, but tend to be varied in effect of open or close-lid container conditions at different binder contents and pressing temperatures. Physical and chemical changes of citric acid after expiration were suggested to be analysed in the further study to investigate the changes of bonding mechanism and its relation to the decreased particleboard properties. This might be an alternative utilisation of expired citric acid from food and pharmacy industries.

ACKNOWLEDGMENTS

The authors acknowledged the assistance of Herdin Ade Surya, Dea Sany Granita, Jasmine Amelia Sidik, Nicky Yulainium Hendra Wijayadi, Fanria Brawnbewer Rumakito, and Adhitya Arvi for data collection in this study.

REFERENCES

Aini EN, Trisatya DR, Santoso A & Dewi GK. 2024. Utilization of expired urea-formaldehyde with the addition of wood vinegar for plywood and particleboard manufacturing. *AIP Conference Proceedings*. 2973: 060010. https://doi.org/10.1063/5.0184556

Aisyadea F, Dewi GK & Widyorini R. 2023. Selected properties of particleboard made from sugar palm (*Arenga pinnata*) dregs. *Journal of the Korean Wood Science and Technology*. 51: 334–344. https://doi.org/10.5658/WOOD.2023.51.5.334

Apelblat A. 2014. *Citric Acid*. Springer International Publishing, Switzerland https://doi.org/10.1007/978-3-319-11233-6

Anosike-Francis EN, Ijeoma Obianyo I, Wasiu Salami O et al. 2022. Physical-Mechanical properties of wood based composite reinforced with recycled polypropylene and cowpea (*Vigna unguiculata* Walp.) husk. *Cleaner Materials*. 5: 100101. https://doi.org/10.1016/j.clema.2022.100101

Arabi M, Haftkhani AR & Pourbaba R. 2023. Investigating the effect of particle slenderness ratio on optimizing the mechanical properties of particleboard using the response surface method. *BioResources*. 18: 2800–2814. https://doi.org/10.15376/biores.18.2.2800-2814

Arendse W & Jideani V. 2022. Storage stability and consumer acceptability of dried apple: impact of citric acid, potassium sorbate and *Moringa oleifera* leaf extract powder. *Foods*. 11: 984. https://doi.org/10.3390/foods11070984

Cheng D, Ding H, Xu B & Li T. 2024. Suitability of thermally modified wood for binderless particleboard production: Effects of temperature and moisture content in hot pressing. *Industrial Crops and Products*. 221*:* 119376. https://doi.org/10.1016/j.indcrop.2024.119376

D'amico S, Hrabalova M, Müller U & Berghofer E. 2010. Bonding of spruce wood with wheat flour glue-effect of press temperature on the adhesive bond strength. *Industrial Crops and Products*. 31: 255–260. https://doi.org/10.1016/j.indcrop.2009.11.001

Dettmer J & Smith GD. 2015. Comparing properties of North American manufactured particleboard and medium density fiberboard - Part I: Particleboard. *BioResources*. 10: 6014–6031. https://doi.org/10.15376/biores.10.3.6014-6031

Dewi GK, Widyorini R & Lukmandaru G. 2022. Application of maltodextrin-based adhesive on particleboard made from *Salacca* frond. *BioResources*. 17*:* 190–206. https://doi.org/10.15376/biores.17.1.190-206

Divkolaei RM, Shalbafan A & Thoemen H. 2024. Influence of panel density on the characteristics of novel multifunctional particleboard. *Wood Material Science & Engineering*: 1–12. https://doi.org/10.1080/17480272.2024.2338202

Fao. 2019. Food and Agriculture Organization of United Nations. Particle Board Production. https://www.fao.org/faostat/en/#data/FO

Fao. 2023. Food and Agriculture Organization of United Nations. Particle Board Production. https://www.fao.org/faostat/en/#data/FO

Ferreira AM, Pereira J, Almeida M et al. 2018. Biosourced binder for wood particleboards based on spent sulfite liquor and wheat flour. *Polymers*. 10: 1070. https://doi.org/10.3390/polym10101070

Flores JA, Pastor JJ, Martinez-Gabarron A, Gimeno-Blanes FJ, Rodríguez-Guisado I & Frutos MJ. 2011. *Arundo donax* chipboard based on urea-formaldehyde resin using under 4 mm particles size meets the standard criteria for indoor use. *Industrial Crops and Products*. 34: 1538–1542. https://doi.org/10.1016/j.indcrop.2011.05.011

Hakim L, Widyorini R, Umemura K, Nugroho Wd & Prayitno TA. 2024. The characteristics of citric acid-bonded composite boards produced from alkaline-modified fibrovascular bundle of *Salacca sumatrana* fronds. *Journal of Natural Fibers*. 21: 2315505. https://doi.org/10.1080/15440478.2024.2315505

Halligain Af & Schniewind AP. 1974. Prediction of particleboard mechanical properties at various moisture contents. *Wood Science and Technology*. 8: 68–78. https://doi.org/10.1007/BF00350644

He Z, Chapital DC, Cheng HN & Modesto OO. 2016. Adhesive properties of water-washed cottonseed meal on four types of wood. *Journal of Adhesion Science and Technology*. 30: 2109–2119. https://doi.org/10.1080/01694243.2016.1175245

Hill CAS. 2006. *Wood Modification Chemical, Thermal and Other Processes*. John Wiley & Sons Ltd, Chichester.

Huang HK, Hsu CH, Hsu PK, Cho YM, Chou TH & Cheng YS. 2022. Preparation and evaluation of particleboard from insect rearing residue and rice husks using starch/citric acid mixture as a natural binder. *Biomass Conversion and Biorefinery*. 12: 633–641. https://doi.org/10.1007/s13399-020-00994-6

Huaxu Z, Hua LS, Tahir PM et al. 2021. Physico-mechanical and biological durability of citric acid-bonded rubberwood particleboard. *Polymers*. 13: 1–12. https://doi.org/10.3390/polym13010098

Hussin MH, Abd Latif NH, Hamidon TS et al. 2022. Latest advancements in high-performance bio-based wood adhesives: A critical review. *Journal of Materials Research and Technology*. 21: 3909–3946. https://doi.org/10.1016/j.jmrt.2022.10.156

Idris NA, Zairun MA, Alias AH, Ibrahim Z & Wahab NA. 2025. Oil palm particleboard: properties and fungi susceptibility. *Journal of Renewable Materials*. 13: 1–10.

https://doi.org/10.32604/jrm.2024.053388

Iskanderani FI. 2008. Physical properties of particleboard panels manufactured from *Phoenix dactylifera*-L (date palm) mid-rib chips using ureaformaldehyde binder. *International Journal of Polymeric Materials and Polymeric Biomaterials*. 57: 979–995. https://doi.org/10.1080/00914030802177404

Islam MN, Rahman F, Das AK & Hiziroglu S. 2022. An overview of different types and potential of bio-based adhesives used for wood products. *International Journal of Adhesion and Adhesives*. 112: 102992. https://doi.org/10.1016/j.ijadhadh.2021.102992

Jis (Japanese Industrial Standard). 2015. *JIS A 5908. Standard Test Methods for Evaluating Particleboard Properties*. Japan Standards Association, Tokyo.

Kim MG, Wan H, No BY & Nieh WL. 2001. Examination of selected synthesis and room-temperature storage parameters for wood adhesive-type urea-formaldehyde resins by 13C-NMR spectroscopy. IV. *Journal of Applied Polymer Science*. 82: 1155–1169. https://doi.org/10.1002/app.1950

Korai H. 2022. Effects of density profile on bending strength of commercial particleboard. *Forest Products Journal*. 72: 85–91. https://doi.org/10.13073/FPJ-D-21-00070

Książek E. 2024. Citric acid: properties, microbial production, and applications in industries. *Molecules* Multidisciplinary Digital Publishing Institute (MDPI), 29: 22. https://doi.org/10.3390/molecules29010022

Kwok K, Mauer LJ & Taylor LS. 2010. Kinetics of moisture-induced hydrolysis in powder blends stored at and below the deliquescence relative humidity: Investigation of sucrose-citric acid mixtures. *Journal of Agricultural and Food Chemistry*. 58: 11716–11724. https://doi.org/10.1021/jf101941e

Laksono AD, Susanto TF, Dikman R, Awali J, Sasria N, & Wardhani IY. 2022. Mechanical properties of particleboards produced from wasted mixed sengon (*Paraserianthes falcataria* (L.) Nielsen) and bagasse particles. *Materials Today: Proceedings*. 65: 2927–2933. https://doi.org/10.1016/j.matpr.2022.01.199

Lee SH, Tahir PM, Lum WC et al. 2020. A review on citric acid as green modifying agent and binder for wood. *Polymers*. 12: 1962. https://doi.org/10.3390/POLYM12081692

Li J, Lei H, Xi X et al. 2023. A sustainable tannin-citric acid wood adhesive with favorable bonding properties and water resistance. *Industrial Crops and Products*. 201: 116933. https://doi.org/10.1016/j.indcrop.2023.116933

Liu R, Liu M, Qu Y, Huang A & Ma E. 2018. Dynamic moisture sorption and formaldehyde emission behavior of three kinds of wood-based panels. *European Journal of Wood and Wood Products*. 76: 1037–1044. https://doi.org/10.1007/s00107-017-1255-y

Malanit P, Barbu MC & Frühwald A. 2009. The gluability and bonding quality of an asian bamboo (*Dendrocalamus asper*) for the production of composite lumber. *Journal of Tropical Forest Science*. 21: 361–368.

Mehrvan K, Jonoobi M, Ashori A & Ahmadi P. 2024. Optimizing paper sludge content and particle size to enhance particleboard properties. *Scientific Reports*. 14(1): 21501. https://doi.org/10.1038/s41598-024-72905-y

Mo X, Hu J, Sun XS & Ratto JA. 2001. Compression and tensile strength of low-density straw-protein particleboard. *Industrial Crops and Products*. 14: 1–9. https://doi.org/10.1016/S0926-6690(00)00083-2

Oh YS. 2020. Effect of job's tears stalk addition in particleboard bonded with urea-formaldehyde resin. *Journal of Tropical Forest Science*. 32: 311-316. https://doi.org/10.26525/jtfs2020.32.3.311

Olawale O, Mathew C, Erinle O, Ajao F & Abayomi S. 2024. Optimization of mixing ratio for the production of particle board from bamboo leaves, saw dust and urea formaldehyde. *Advances in Bamboo Science*. 9: 1–8. https://doi.org/10.1016/j.bamboo.2024.100108

Patel DS, Dholakia AB, Patel VC & Patel AJ. 2024. Development of defatted soy flour and casein-based bio adhesives for wood joint. *Journal of Tropical Forest Science*. 36: 274–279. https://doi.org/10.26525/jtfs2024.36.3.274

Pizzi A, Papadopoulos AN & Policardi F. 2020. Wood composites and their polymer binders. *Polymers*. 12: 1–27. https://doi.org/10.3390/POLYM12051115

Pędzik M, Janiszewska D & Rogoziński T. 2021. Alternative lignocellulosic raw materials in particleboard production: A review. *Industrial Crops and Products*. 174: 114162. https://doi.org/10.1016/j.indcrop.2021.114162

Radabutra S, Khemthong P, Saengsuwan S & Sangya S. 2020. Preparation and characterization of natural rubber bio-based wood adhesive: Effect of total solid content, viscosity, and storage time. *Polymer Bulletin*. 77: 2737–2747. https://doi.org/10.1007/s00289-019-02881-1

Scharf A, Sandberg D & Jones D. 2024. The influence of chemical content and pressing temperature on the properties of citric acid-bonded particleboards from softwood sawmilling residues. *Wood Material Science and Engineering*. 19: 987–992. https://doi.org/10.1080/17480272.2024.2367608

Simer K, Kaljuvee T, Christjanson P & Pehk T. 2005. Changes in Curing Behaviour of Aminoresins during Storage. *Journal of Thermal Analysis and Calorimetry*. 80: 123–130. http://doi.org/10.1007/s10973-005-0623-8

Simmons JE. 1995. Storage in Fluid Preservatives. In Rose CL, Hawks CA, Genoways HH (eds.). *Storage of Natural History Collections: A Preventive Conservation Approach*. Society for the Preservation of Natural History Collections, Iowa City.

Bps-Statistic Indonesia. 2020. *Statistic of Forestry Production 2019*. Statistic Indonesia, Jakarta. https://www.bps.go.id/en/publication/2020/11/27/969702bd7c227b224b293795/statistics-of-forestry-production-2019.html

Bps-Statistic Indonesia. 2024. *Statistic of Forestry Production 2023*. Statistic Indonesia, Jakarta. https://www.bps.go.id/en/publication/2024/07/26/3a3802857697

0e086c1cf32f/statistics-of-forestry-production-2023.html

Stoeckel F, Konnerth J & Gindl-Altmutter W. 2013. Mechanical properties of adhesives for bonding wood-A review. *International Journal of Adhesion and Adhesives.* 45: 32–41. https://doi.org/10.1016/j.ijadhadh.2013.03.013

Tabor D. 1951. *The hardeness of metal.* Clarendon Press, Oxford.

Umemura K, Hayashi S, Tanaka S & Kanayama K. 2017. Changes in physical and chemical properties of sucrose by the addition of ammonium dihydrogen phosphate. *Journal of The Adhesion Society of Japan* 53: 112-117. https://doi.org/10.11618/adhesion.53.112

Umemura K, Ueda T & Kawai S. 2012. Characterization of wood-based molding bonded with citric acid. *Journal of Wood Science.* 58: 38–45. https://doi.org/10.1007/s10086-011-1214-x

Wardani L, Massijaya MY, Hadi YS, Darmawan IW. 2015. Kualitas papan zephyr pelepah sawit dan papan komposit komersial sebagai bahan bangunan (in Bahasa). *Jurnal Teknik Sipil.* 22*:* 79–86.

Wibowo ES, Kusumah SS, Subyakto & Umemura K. 2021. Modification of novel bio-based adhesive made from citric acid and sucrose by $ZnCl_2$. *International Journal of Adhesion and Adhesives.* 108: 1–12. https://doi.org/10.1016/j.ijadhadh.2021.102866

Widyorini R, Umemura K, Isnan R, Putra DR, Awaludin A & Prayitno TA. 2016. Manufacture and properties of citric acid-bonded particleboard made from bamboo materials. *European Journal of Wood and Wood Products.* 74: 57–65. https://doi.org/10.1007/s00107-015-0967-0

Widyorini R, Umemura K, Kemalasari DS, Dewi GK & Nugroho WD. 2019. Effect of citric acid content and extractives treatment on the manufacturing process and properties of citric acid-bonded *Salacca* frond particleboard. *BioResources.* 14: 4171–4180. https://doi.org/10.15376/biores.14.2.4171-4180

Widyorini R, Umemura K, Septiano A, Soraya DK, Dewi GK & Nugroho WD. 2018. Manufacture and properties of citric acid-bonded composite board made from *Salacca* frond: effect of maltodextrin addition, pressing temperature, and pressing method. *BioResources.* 13: 8662–8676. https://doi.org/10.15376/biores.13.4.8662-8676

Zeleniuc O, Brenci L-M, Cosereanu C & Fotin A. 2019. Influence of adhesive type and content on the properties of particleboard made from sunflower husks. *BioResources.* 14*:* 7316–7331. https://doi.org/10.15376/biores.14.3.7316-7331

Zhang Q, Pizzi A, Lei H, Xi X, Cao M & Cao L. 2023. MALDI ToF investigation of the reaction of soy protein isolate with glutaraldehyde for wood adhesives. *Journal of Renewable Materials.* 11: 1439–1450. https://doi.org/10.32604/jrm.2022.023535