The Effect of UF Addition on the Properties of Citric Acid-Starch Bonded Particleboard

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ABSTRACT

Citric acid and starch have attracted considerable interest as potential modifiers for urea-formaldehyde (UF) adhesives, owing to their capability to improve performance and promote environmental sustainability. This study examines the impact of varying urea-formaldehyde (UF) content added to a citric acid-tapioca starch adhesive system on the performance of rubberwood particleboard. The total resin content was set at 10%, with the adhesive composed of a mixture of citric acid (CA) and tapioca starch in a 75/25 ratio. UF was added to the adhesive in three different formulations: 2%, 5%, and 10% of the total resin content. The resulting particleboards were then tested for thickness swelling (TS), water absorption (WA), internal bond strength (IB), modulus of elasticity (MOE), and modulus of rupture (MOR). The findings indicate that increasing UF content improves the mechanical properties, such as the modulus of rupture (MOR) and modulus of elasticity (MOE). However, higher UF content also negatively impacts the physical properties, resulting in greater thickness swelling (TS) and water absorption (WA), which may compromise the overall dimensional stability and internal bond (IB) strength. The study concludes that UF content of around 5% offers an optimum property, achieving strong mechanical performance (IB = 0.207 N/mm², MOR = 8.35 MPa, and MOE = 1645.15 MPa) while managing physical properties within acceptable ranges (TS = 17.5120.15%, WA = 87.8793.43%, Density = 565.36 kg/m³).

Keywords: dimensional stability, mechanical stability, particleboard, rubberwood, starch, UF

INTRODUCTION

Particleboards are essential to various industrial sectors, particularly furniture manufacturing and construction, due to their affordability and adaptability. These engineered wood products are produced by bonding wood particles together with a resin adhesive through heat and pressure, making them suitable for applications ranging from kitchen cabinetry to flooring substrates. Rubberwood, among the woods commonly used, has become especially significant due to its renewable nature and abundant supply in tropical areas (Hashim et al. 2015). Rubberwood has become a key raw material for Malaysia's composite panel industry, with waste from furniture and lumber production now serving as the primary source for these products (Amini et al. 2013).

The quality of adhesives plays a critical role in determining the properties of particleboard, such as its mechanical strength and resistance to environmental factors like moisture (Baharuddin et al. 2023). Formaldehyde-based adhesives are extensively utilised in the production of particleboard as its primary binder due to its rapid drying time, transparent appearance, and economical price such as urea formaldehyde (UF) (Ashori & Kuzmin 2024). This cost advantage helps in maintaining the affordability of particleboard products, contributing to its widespread use in various applications (Owodunni et al. 2020). The use of UF binders in wood composite boards presents two significant drawbacks: firstly, the production process results in substantial formaldehyde emissions, and secondly, the boards are susceptible to damage from moisture exposure and swelling, as UF-based adhesives have limited water resistance, leading to potential dimensional instability and compromised structural integrity during use (Pizzi et al. 2020). However, the environmental impact of formaldehyde emissions (Yang & Rosentrater 2020) and low dimension stability (Özgenç Keleş & Nemli 2020) from UF resins has propelled the search for more eco-friendly alternatives.

Citric acid and starch have garnered significant attention as potential modifiers for urea-formaldehyde (UF) adhesives due to their ability to enhance both performance and environmental sustainability. Citric acid, a natural organic acid, can improve the bonding performance of UF adhesives by introducing additional cross-linking sites, thereby enhancing water resistance and reducing formaldehyde emissions (Lee et al. 2020, Dunky 2021, Hussin et al. 2022). Meanwhile, starch, a biodegradable polysaccharide, offers a sustainable and renewable modification to the adhesive matrix, potentially improving dimensional stability and mitigating the brittleness of the resulting composite (Maulana et al. 2022). Research has demonstrated that starch-based adhesives can provide satisfactory bonding strength and notable environmental benefits when incorporated into wood composites (Watcharakitti et al. 2022). The integration of citric acid and starch is a promising approach to enhance the mechanical and dimensional stability of rubberwood particleboard (Widyorini et al. 2017). A research added corn, ganyong (Canna edulis Ker-Gawl), and garut (Maranta arundinacea L.) starches to citric acid as an adhesive for particleboard made from Petung (Dendrocalamus sp.) bamboo particles (Widyorini et al. 2017). In a study, 2% urea-formaldehyde (UF) was added to a citric acid and corn starch adhesive formulation for manufacturing rubberwood particleboard, resulting in enhanced internal bonding (IB) strength and modulus of rupture (MOR), both of which met standard requirements (Amini et al. 2020).

This study seeks to fill the research gap concerning the acid-starch adhesives and UF especially on tapioca starch, with a focus on the suitability of formulation incorporates small amounts of UF into the adhesive formulation of citric acid and tapioca starch. By examining the effect of UF addition on the properties of citric acid-starch bonded rubberwood particleboard, these findings can be used to deduce the formulation of the adhesives, supporting the goal of creating a high-performance composite suitable for diverse applications while addressing environmental and health concerns linked to traditional adhesives.

MATERIALS AND METHODOLOGY

Preparation of materials

The particles from the Rubberwood at the age of around 35 years were procured from HeveaBoard Berhad, located in Gemas, Negeri Sembilan. These particles were subsequently dried in a laboratory oven to a moisture content of 3% in preparation for particleboard production. The adhesives utilised in this study comprised urea-formaldehyde (UF) resin, citric acid (CA) and tapioca starch. The UF resin, with a solid content ranging from 60% to 65%, was sourced from Aica Chemicals (M) Sdn. Bhd in Senawang. Citric acid, obtained in powdered form from Evergreen Engineering & Resources in Semenyih, Selangor, was prepared into a 60% solid solution using distilled water to serve as a binder in the manufacturing process. Tapioca starch was supplied by Chop Lee Seng in Serdang, Selangor and sourced from a local grocery store without a specific brand designation.

Preparation of adhesive solution

The adhesive solutions were prepared by initially mixing citric acid (CA) and tapioca starch in a 75:25 weight ratio. This mixture was dissolved in hot water at 70°C, with a solid content of 60 wt.%. Subsequently, urea-formaldehyde (UF) resin was added to the citric acid-tapioca starch solution in varying amounts of 0%, 2%, 5%, and 10% by weight, based on the total adhesive content. The UF resin was thoroughly incorporated to ensure homogeneity before being utilized in the production of rubberwood particleboard. A set of particleboards made with only urea-formaldehyde (UF) resin adhesive was used as the control.

Table 1. Types of binder prepared for the manufacturing of particleboard.

Label	Binder
Control (UF)	UF
0%	CA + tapioca starch
2%	98% CA + tapioca starch, 2% UF
5%	95% CA + tapioca starch, 5% UF
10%	90% CA + tapioca starch, 10% UF

Production of particleboard

Before particleboard fabrication, rubberwood particles were dried to a moisture content of 3% at a temperature of 103 ± 2°C. Particleboards, with dimensions of 340 mm × 340 mm × 12 mm and a target density of 700 kg/m^3, were produced using 10 wt% adhesive based on the dry weight of the rubberwood particles. The production process began with rubberwood particles being added to a blender, where the adhesive was simultaneously sprayed during mixing. After blending, the resin-coated particles were transferred into a wooden mold to form a mat, which was then compacted by pre-pressing. The mat was subsequently hot-pressed at 180°C for 10 minutes under a pressure of 100 bar. A hot-pressing temperature of 180°C was chosen based on a study demonstrating that using citric acid as an adhesive for particleboard achieves optimal performance at this temperature, yielding superior bonding strength, which improves as the pressing temperature increases (Cahyono and Syahidah, 2019). Following hot-pressing, the particleboards were conditioned for 7 days in a controlled environment at 23 ± 2°C and 65 ± 5% relative humidity to ensure stabilization prior to property evaluation. This process was repeated for each adhesive formulation, 3 particleboards were produced for each variable, resulting in a total of 15 particleboards.

Properties Evaluation

Dimensional Stability

A) Thickness Swelling (TS)

Thickness swelling (TS) and Water absorption (WA) tests were conducted according to the methods reported by other research (Odeyemi et al., 2020). The particleboard samples were cut into 50 mm × 50 mm squares. The thickness of each sample was precisely measured at the intersection of the diagonals using vernier calipers with an accuracy of 0.01 mm. Subsequently, the samples were immersed in clean water, and their thickness was re-measured after 2 hours and 24 hours of submersion to assess thickness swelling. Thickness swelling was then calculated using the following formula:

$$TS\ (\%) = (T_2 - T_1)/T_1 \times 100$$

where,
T_1: Initial thickness of sample before immersion (mm)
T_2: Thickness of sample after immersion for 2 hours/24 hours (mm)

B) Water Absorption (WA)

The particleboard samples, cut into 50 mm × 50 mm squares, were initially weighed before immersion. The weight gain of these samples was then recorded after 2 hours and 24 hours of submersion. The percentage increase in weight, representing water absorption, was calculated using the following formula:

$$WA\ (\%) = (W_2 - W_1)/W_1 \times 100$$

where,
W_1: Initial weight of sample before immersion (g)
W_2: Weight after immersion for 2 hours/24 hours (g)

Mechanical properties

Mechanical properties such as modulus of rupture (MOR), modulus of elasticity (MOE) and internal bonding (IB) were determined in accordance with JIS A 5908: 2003 standards using universal testing machine INSTRON 5582, 100kN. For MOR and MOE, the particleboard was cut into 200mm x 50mm squares. Results of MOR and MOE were obtained directly from the universal testing machine. For IB, the particleboard was cut into 50mm x 50mm squares. The results of IB were calculated using the following formula:

$$\text{Internal bond } (N/mm^2) = P'/\ 2bL$$

Where,
P’: maximum load (N) at the time of failing force
b: width (mm) of sample
L: length (mm) of sample

RESULT AND DISCUSSION

Dimensional Stability

The data presented in the Table 2 showed the effects of varying concentrations of UF on the water absorption properties of rubberwood particleboards. In this research, the density of the manufactured particleboards ranged from 585.18 to 633.21 kg/m³. The parameters investigated thickness swelling (TS) after 2 and 24 hours (TS-2h, TS-24h) and water absorption (WA) after 2 and 24 hours (WA-2h, WA-24h), alongside the density measurements of the boards.

Dimensional stability denotes a material's capacity to retain its original dimensions under varying moisture and temperature conditions. For rubberwood particleboards, this stability is chiefly evaluated through thickness swelling (TS) and water absorption (WA) measurements. The provided data demonstrates that the addition of UF-CA-starch adhesive significantly influences the dimensional stability of rubberwood particleboards. The control group, bonded with UF, with thickness swelling of 29.43% at 2 hours and 44.08% at 24 hours. Compare to the particleboard bonded with CA and tapioca starch, when 2% UF concentration is applied, thickness swelling significantly decreases to 19.02% at 2 hours and 23.55% at 24 hours, indicating a marked improvement in dimensional stability. This trend continues with a 5% UF concentration, yielding TS-2h at 17.51% and TS-24h at 20.15%, further showcasing enhanced stability. However, at a 10% UF concentration, thickness swelling rises to 29.22% at 2 hours and 33.02% at 24 hours, suggesting a deterioration in dimensional stability.

Regarding water absorption, the control group exhibited a value of 90.57% at 2 hours and 114.92% at 24 hours, indicating poor dimensional stability due to excessive water uptake, which is slightly higher than that of the particleboard bonded with CA and tapioca starch. At a 2% UF concentration, water absorption significantly decreased to 76.70% at 2 hours and 90.95% at 24 hours, demonstrating enhanced water resistance and improved dimensional stability. However, at a 5% UF concentration, water absorption rates increased to 87.87% at 2 hours and 93.43% at 24 hours. Although these values are still superior to those of the control group, they suggest a plateau in the improvement of water resistance. Conversely, at a 10% UF concentration, water absorption rates further increased to 90.28% at 2 hours and 95.12% at 24 hours, indicating a decline in water resistance and, consequently, reduced dimensional stability.

The observed reductions in thickness swelling and water absorption at lower UF concentrations (2% and 5%) indicate a substantial improvement in both dimensional stability and water resistance, likely attributable to the cross-linking within the citric acid and starch. Wood composites containing cross-linked starch exhibited greater resistance to water uptake due to the reduction of hydroxyl groups in native starch through a condensation reaction and the formation of a three-dimensional network during the curing process (Kaith et al. 2010).

The increased thickness-swelling and water absorption observed at the highest UF concentration (10%) might be attributed to several factors such as over saturation of the adhesive, potential interference in the bonding mechanism, or the physical properties of the adhesive itself altering under higher concentrations. As stated by Stefanowski et al. (2018), the characteristic of UF which has poor tolerance towards the water which causes hydrolysis easier. At higher concentrations, the viscosity of the adhesive mixture might increase, potentially affecting its ability to penetrate and uniformly coat the wood particles. Incomplete or uneven distribution of the adhesive could lead to weak spots within the board where moisture can more easily penetrate and cause swelling (Frihart & Hunt 2010). In addition, higher concentrations of additives might alter the physical properties of the adhesive layer, such as its flexibility, porosity, or thickness, each of which could impact water resistance and dimensional stability. For example, a thicker adhesive layer might be more prone to cracking or might not cure uniformly, thereby allowing more moisture intrusion (Frihart 2005). This implies that while the additive enhances certain properties, there is an optimal concentration beyond which the benefits do not continue to accrue and may in fact reverse.

Additionally, density plays a critical role in influencing both water absorption and thickness swelling in wood composites. Typically, as the density of wood composites increases, thickness swelling also rises. This is primarily due to the release of residual compressive stresses that are introduced during the hot-pressing process of the board. Upon exposure to elevated humidity or liquid water, these stresses can cause the material to expand, resulting in a phenomenon referred to as "spring back" (Amini et al. 2020) This effect describes a form of non-recoverable thickness swelling, whereby the wood composite irreversibly increases in thickness after encountering moisture (Kelly 1977).

In summary, the analysis of thickness, swelling and water absorption demonstrates that these findings highlight the potential of the UF-Citric Acid-Starch adhesive to improve the performance of rubberwood particleboards, although the optimal concentration for achieving the best balance of properties appears to be below 10%. An optimal balance of adhesive content is crucial for achieving desired dimensional stability and minimizing water absorption in rubberwood particleboards.

Table 2. Density, water absorption and thickness swelling of particleboard bonded with different UF content.

Type	TS-2h (%)	TS-24h (%)	WA-2h (%)	WA-24h (%)	Density (kg/m^3)
Control (UF)	29.43 ± 3.77b	44.08 ± 4.08c	90.57 ± 3.74c	114.92 ± 4.47d	692.49 ± 11.76a
0%	45.92 ± 5.54c	50.21 ± 5.48d	98.29 ± 5.33d	105.51 ± 8.68c	692.53 ± 19.54a
2%	19.02 ± 5.14a	23.55 ± 7.62a	76.70 ± 4.93a	90.95 ± 5.72a	585.18 ± 43.29c
5%	17.51 ± 1.59a	20.15 ± 1.58a	87.87 ± 3.87b	93.43 ± 2.87b	565.36 ± 13.92c
10%	29.22 ± 3.61b	33.02 ± 4.37b	90.28 ± 3.90c	95.12 ± 2.39b	633.21 ± 33.33b

Note: Values after ± are standard deviations
Within the same column, means followed by the different letters a, b, c and d are significantly different at $p \leq 0.05$.

Mechanical properties

The internal bond (IB) strength of the particleboard samples was measured to evaluate the effect of varying urea-formaldehyde (UF) content. The control sample (UF) exhibited the lowest IB strength at 0.15 MPa, precisely meeting the minimum standard requirement (0.15 MPa). Compared to particleboards bonded solely with CA and tapioca starch, the inclusion of UF resulted in a higher IB strength. Incorporation of 2% UF significantly increased the IB strength to 0.217 MPa, indicating a strong adhesive effect. However, increasing the UF content to 5% resulted in a slight decrease in IB strength to 0.207 MPa. This trend continued with the 10% UF sample, which showed a further decline in IB strength to 0.175 MPa. The results indicated that although low concentrations of urea-formaldehyde (UF) improve internal bonding, higher UF levels tend to adversely affect the internal cohesion of the particleboard, aligning with the findings reported by Prasetiyo et al (2019). The modulus of rupture (MOR) results revealed that the bending strength of the particleboard samples improved with increasing UF content. The control sample had a MOR of 9.27 MPa. The particleboard bonded with CA with tapioca starch had a MOR of 7.07 MPa. When 2% UF was added, the MOR increased to 7.84 MPa. Further increase in UF content to 5% resulted in the highest MOR of 8.35 MPa. The 10% UF sample also showed a high MOR of 8.38 MPa, indicating that higher UF content positively influences the bending strength. However, the increase in MOR between 5% and 10% UF is minimal. The modulus of elasticity (MOE) was also affected by the UF content in the particleboard. The control sample had an MOE of 1374.59 MPa. With the addition of 2% UF, the MOE increased to 1529.88 MPa, demonstrating improved stiffness. The trend continued with 5% UF, resulting in an MOE of 1645.15 MPa. The 10% UF sample showed the highest MOE at 1663.79 MPa. These results indicate that UF content positively correlates with the stiffness of the particleboard, with the greatest improvements observed at higher UF levels.

The findings from this study reveal that the addition of UF resin significantly enhances the mechanical properties of particleboard compared with the particleboard bonded with only CA and starch, particularly in terms of IB, MOR, and MOE. The initial increase in IB strength with 2% UF suggests that UF acts as an effective adhesive at lower concentrations. However, the subsequent decrease in IB strength at higher UF levels (5% and 10%) may be attributed to weakening of the internal structure. The excess UF resin may form a brittle matrix that lacks the flexibility to withstand internal stresses (Stoeckel et al. 2013, Amini et al. 2020, LEE et al. 2020), thereby reducing the internal bond (IB) strength.

The progressive increase in MOR and MOE with higher UF content indicates that UF resin contributes to the overall composite strength and rigidity (Prasetiyo et al. 2019). Apart from control samples, the highest improvements in bending strength and elasticity were observed with 5% and 10% UF, suggesting that these concentrations are optimal for maximizing mechanical performance without compromising internal bond strength.

As a result, while UF resin enhances the mechanical properties of particleboard, there is a critical balance to be maintained. Optimal improvements were observed with 5% UF content, beyond which the benefits in IB strength diminish.

Table 3. Internal bond and bending strength of particleboard bonded with different UF content.

Type	IB (MPa)	MOR (MPa)	MOE (MPa)
Control (UF)	0.15± 0.06d	9.27 ± 0.19a	1543.01 ± 68b
0%	0.19 ± 0.05b	7.07 ± 0.33d	1374.59 ± 45c
2%	0.217 ± 0.06a	7.84 ± 0.63c	1529.88 ± 36b
5%	0.207 ± 0.04a	8.35 ± 0.82b	1645.15 ± 62a
10%	0.175 ± 0.02c	8.38 ± 0.65b	1663.79 ± 75a

Note: Values after ± are standard deviations
Within the same column, means followed by the different letters a, b, c and d are significantly different at $p \leq 0.05$.

CONCLUSION

The addition of UF in the adhesive of CA-starch could improve the denominational stability and the mechanical properties of particleboard. The incorporation of UF-CA-starch adhesive significantly improves the dimensional stability of rubberwood particleboards at lower concentrations (2% and 5%). The observed reductions in thickness swelling and water absorption at these concentrations suggest enhanced resistance to moisture-induced dimensional changes. The results indicate that the 2% UF addition yields the highest internal bond (IB) strength, while the 5% UF addition provides an optimal balance between mechanical properties and dimensional stability in UF-CA-starch bonded particleboards. The results demonstrate the potential of UF-CA-starch adhesives to offer a more environmentally sustainable alternative to traditional adhesives. Future research should explore further optimization of UF ratios or investigate the potential benefits of combining UF with other additives to further enhance the overall performance of particleboards.

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