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Adhesive Characterization of *Acacia mangium* Bark Extract and Its Effects on Properties and Formaldehyde Emissions of Rubberwood Particleboard

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ABSTRACT

This study evaluated the feasibility of using *Acacia mangium* bark extract (BE) as a bio-based, formaldehyde-free adhesive for rubberwood particleboard, focusing on the effects of two catalysts: sodium hydroxide (alkaline) and boric acid (acidic). Gel time, viscosity, thermal behavior, mechanical performance, and formaldehyde emissions were assessed. The uncatalyzed BE adhesive showed a gel time of 29.59 min, which decreased to 6.44 min with sodium hydroxide (BE–NaOH adhesive) and 4.22 min with boric acid (BE–Boric adhesive), highlighting the catalytic effect. Differential scanning calorimetry revealed higher curing enthalpy for BE–NaOH adhesive, while thermogravimetric analysis indicated enhanced thermal stability under alkaline conditions. The viscosity of BE–NaOH adhesive was comparable to commercial urea formaldehyde (UF) adhesive. Particleboards (density 0.65 g/cm^3) bonded with BE–NaOH, BE, or BE–Boric adhesive met the JIS A 5908:2015 Type 8 internal bond (IB ≥ 0.15 N/mm^2) requirement. Boards bonded with BE achieved a modulus of rupture of 8.25 N/mm^2, meeting the Type 8 threshold (≥ 8 N/mm^2), while those with BE–NaOH adhesive showed 7.61 N/mm^2, slightly below it. Formaldehyde emissions were very low (0.006–0.058 mg/L), achieving the F**** rating under JIS A 5908:2015. However, high thickness swelling and water absorption indicated limited water resistance, and overall mechanical properties remained lower than with UF/melamine urea formaldehyde (MUF) adhesives. Direct mechanical performance comparison with UF/MUF was not possible due to the lack of molar ratio data. These findings suggest BE adhesives are a promising sustainable alternative, though further improvements in water resistance and mechanical performance are needed.

Keywords: bark extracts, rubberwood, particleboard, non-formaldehyde adhesives

1. INTRODUCTION

Wood-based panel composites, namely plywood, particleboard, or medium density fiberboard, are widely used in residences. They are commonly used in furniture, flooring, and interior and exterior joinery. In Thailand, rubberwood is the main raw material for wood composite panels, including particleboard, medium-density fiberboard, oriented strand board, and plywood. In 2023, Thailand is expected to be the third largest exporter of particleboard in the world, with an export volume of 6,995,030 m^3 (https://www.fao.org/faostat/en/#data/FO/

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visualize). Formaldehyde-based adhesives such as urea formaldehyde (UF), melamine formaldehyde, melamine-urea formaldehyde (MUF), and phenol formaldehyde (PF), are the generally used binders in these composites. The selection of adhesive for particleboard or medium density fiberboard depends on several aspects, such as the required strength properties, the estimated moisture resistance, the expected formaldehyde emission class, and the production cost of the finished product. In the processing, fast curing is preferred, and water solubility would facilitate the adjustment of viscosity (Solt *et al*., 2019).

Formaldehyde emissions from wood-based panels in buildings are known to be seasonally variable (Liang *et al*., 2021) and represent a major contribution to indoor air pollutants (Böhm *et al*., 2012; Huang *et al*., 2022; Zhou *et al*., 2022). Formaldehyde, classified as a human carcinogen by the International Agency for Research on Cancer (IARC), can be released from wood products in indoor environments and potentially lead to the sick building syndrome (SBS; Hu *et al*., 2022). The SBS symptoms fall into 3 categories, including general symptoms (fatigue, nausea/dizziness, difficulty concentrating, heavy-headedness, and headache), mucous membrane irritation (irritation of the eyes; irritated, stuffy, or runny nose; and cough), and dry skin symptoms (dry throat and hoarseness, dry or flushed facial skin, scaly/itchy scalp or ears, and dry hands; Suzuki *et al*., 2021).

Several investigations have been carried out to decrease the release of formaldehyde, for example by lowering the formaldehyde to urea molar ratio, by using additives (Atar *et al*., 2014) and by applying formaldehyde scavengers (Ghani *et al*., 2018). A coating on wood-based material can act as a diffusion barrier, hindering formaldehyde release from the base material (Pibiri *et al*., 2020). Bio-based adhesives with components such as cellulose, protein, lignin, and tannin demonstrate great potential for bonding specially engineered wood products (Islam *et al*., 2022). The panels bonded with modified lignin resins present lower formaldehyde emissions than those bonded with PF resin (Younesi-Kordkheili, 2022). Condensed tannins extracted from mangrove bark (*Rhizophora apiculata*) have been used as binder in plywood (Irman *et al*., 2022). An adhesive composed of oxidized corn starch and urea-based adhesive can be used with traditional MUF in a hybrid adhesive system for producing particleboards with low formaldehyde content (Oktay *et al*., 2021). When particleboards were prepared with tannin adhesive extracted from chestnut shell, chestnut bur, and eucalyptus bark, they were classified as E0 because of the low formaldehyde emissions (Santos *et al*., 2017). Therefore, developing resin formulations with lower toxicity from renewable resources is beneficial, especially for reducing formaldehyde exposure.

Acacia mangium is an economic fast-growing tropical plantation tree used for furniture, cabinets, floors, particleboard, plywood, veneer, fence posts, firewood, and charcoal. However, *Acacia mangium* bark has not been properly utilized. Currently, the bark is incinerated or landfilled. Mainly, it is used as a low-cost fuel in sawmills.

Acacia mangium bark is mainly composed of holocellulose (36.2%), cellulose (22.4%), lignin (14.7%), and hot water-soluble extractives (17.4%; Hoong *et al*., 2011). Yingprasert *et al*. (2023) found that *Acacia mangium* bark extracts (BEs) contained at least 70 chemical components. The main constituents included 1,3,5-cycloheptatrienemethanol, nitromethane, 1,2,3-propanetriol, hexadecanoic acid, ethyl ester, acetol, phenol, and 2-methoxy and methyl glycolates. In general, the pH levels show that the *Acacia mangium* bark is more acidic (pH 4.5) than the wood fraction (pH 6.1). Condensed tannin, the component of BEs can react with formaldehyde and form methylene and oxymethylene bridges, which contribute to network structures that are stable against hydrolysis. The Stiasny number serves as an indicator of a tannin's ability to react with formaldehyde. According

to Hendrik *et al.* (2019), condensed tannin extracted from *Acacia mangium* exhibited a Stiasny number of 47.22%, containing approximately 49.08% phenolic constituents and having an average molecular weight of 4,745. The extraction of *Acacia mangium* bark using aqueous sulfite–carbonate solution produced relatively high tannin yields with a high Stiasny number, which is suitable for plywood adhesives. It was reported that extracts from *Acacia. mangium* tree bark are rich in phenolic compounds and have the potential to replace conventional PF adhesive used in plywood manufacturing (Hoong *et al*., 2011). The phenolic constituents of larch and valonia tannins enable their reaction with formaldehyde, supporting their potential application in wood adhesive formulations (Li *et al*., 2016a). *Acacia mangium* bark powder, when added to phenol-formaldehyde resin, enhances the condensation of hydroxymethyl groups, promoting the formation of methylene bridges and dimethylene ether bridges, thus increasing the bond strength of plywood pressed at 110℃ (Miyazaki and Hirabayashi, 2011). The properties of particleboard made with a synthesized bio-based resin containing cornstarch, Mimosa tannin, sugar, and citric acid, generally meet the requirements for panels used in dry medium interior fittings (P2) according to European norms EN 312 (Oktay *et al*., 2021). Maritime pine tannin resin (10% by weight of solids content in adhesive/dry weight of wood) was used to form pine wood particleboard (density 670 kg/m^3) under 220℃ for 7.5 minutes with a pressure cycle of 2.7 MPa/1.47 MPa/0.5 MPa for 2 min/2.5 min/3 min, respectively. It was found that the internal bond (IB) of particleboard made with this adhesive (0.64 MPa) was comparable to that obtained with UF resin (0.72 MPa; Ndiwe *et al*., 2019). A sodium alginate (SA)-corn-starch-mimosa tannin (CSMT) wood adhesive was prepared and tested for potential particleboard applications. The mechanical results obtained showed a significant degradation at low and high contents of SA as a partial substitution in CSMT. However, CSMT/SA (99.5/0.5; w/w) demonstrated an excellent mechanical and physical performance (Boussetta *et al*., 2021).

The primary aim of this research was to investigate the feasibility of using *Acacia mangium* BE as a bio-based and formaldehyde-free adhesive to produce rubberwood particleboard. In the initial phase, the effect of pH on the synthesis of the adhesive was investigated without the addition of any hardeners. Two alternative types of catalysts, an alkaline catalyst (sodium hydroxide) or an acidic catalyst (boric acid), were added during the preparation of the adhesives. The properties of the adhesives, including solids content and viscosity, were investigated. The physical and mechanical properties of the panels made with the alternative adhesive formulations were evaluated. The formaldehyde emissions from the boards were also determined. Finally, the properties of particleboards using *Acacia mangium* BEs as adhesives were compared with those made with commercial UF and MUF adhesives. The findings from this study are expected to provide valuable insights into the potential of condensed tannin-rich BEs for sustainable particleboard production and to inform future research on the industrial application of bio-based adhesives.

2. MATERIALS and METHODS

2.1. Materials

2.1.1. *Acacia mangium* bark

Acacia mangium bark was collected from an area within the Prince of Songkla University, Surat Thani Campus, Surat Thani province, Thailand. The age and diameter at the bottom part of the trees were approximately 13 to 18 years and 20 to 30 cm, respectively. Bark at the butt portion of stems was gathered using traditional hand-cutting tools.

2.1.2. Rubberwood particles

Rubberwood particles were supplied by S.P.B. Panel

Industries, a local particleboard company in Ban Na Doem, Surat Thani province, Thailand, for particleboard manufacturing in the laboratory. The particles were dried in an oven at 103 ± 2℃ for 24 hours. The moisture content of dried particles was approximately 2%–3%.

2.1.3. Urea formaldehyde and melamine urea formaldehyde adhesives

UF and MUF adhesives were provided by Aica Hatyai, located in Songkhla province, Thailand.

2.2. *Acacia mangium* bark extract adhesive formulation

The bark particles were cut to smaller size and extracted with distilled water, following with the method reported by Yingprasert *et al*. (2014). The bark was ground into particles passing through 40 mesh sieve and then extracted with distilled water (1:15 w/v) at 100℃ for 4 h. Approximately 100 g of ground bark and 1,500 mL of distilled water were used for each extraction. Initially, the solution was filtered through Whatman No.1 filter paper, followed by filtration through sintered glass (40–100 μm). The extract solution was then evaporated to retain its solids, at 50℃ in an oven until constant weight. The extraction yield was 12.04 ± 1.71% of the initial dry weight of the bark. The Stiasny number was 86.16 ± 2.63% and other phenolic material that can be reacted with formaldehyde was 12.48 ± 0.54% (w/w) of bark. The BEs contained 17.75 ± 2.08 mg catechin equivalents (CE) per g of bark as condensed tannin content (Yingprasert *et al*., 2014). The *Acacia mangium* BE adhesive was formulated by dissolving 50 g of the *Acacia mangium* BE in 750 mL of distilled water (1:15 w/v) in a five-neck flask reactor maintained at 80℃ for 180 min under continuous stirring at 360 rpm. The second and third adhesive formulations were prepared following the same initial procedure as for the BE adhesive. In brief, 50 g of *Acacia mangium* BE was dispersed in 750 mL of distilled water (1:15 w/v) in a five-neck flask reactor and maintained at 80℃ for 90 min under continuous stirring at 360 rpm. Subsequently, 40% NaOH solution was added to the second formulation to adjust the pH to 10 (designated as BE-NaOH), while boric acid was introduced into the third formulation to adjust the pH to 3 (designated as BE-Boric). Both mixtures were further stirred for an additional 90 min under the same conditions, resulting in a total reaction time of 180 min, consistent with the preparation procedure of the BE adhesive. The synthesis reactions for all adhesive formulations were terminated by rapidly cooling the mixtures to room temperature after 180 min of reaction at 80℃.

The selection of sodium hydroxide (NaOH) and boric acid as catalysts in this study was primarily based on two considerations. First, we aimed to explore the effects of acidic and alkaline conditions on the synthesis and performance of the BE-based adhesive, given that pH is known to significantly influence the reactivity of phenolic compounds such as condensed tannins. Second, both NaOH and boric acid have been reported in previous studies to be compatible with tannin-based adhesive systems and are commonly used in wood adhesive research as pH modifiers. Sodium hydroxide was selected to create an alkaline environment that enhances the nucleophilic reactivity of phenolic hydroxyl groups, potentially promoting polymerization. In contrast, boric acid was used to investigate how an acidic medium would affect the adhesive characteristics, as it may also act as a mild crosslinking agent. Therefore, the catalysts were chosen both for their chemical relevance and to represent contrasting pH conditions for a comparative evaluation.

2.3. Differential scanning calorimetry

Curing properties of adhesives were tested using differential scanning calorimetry (DSC; Differential Scanning Calorimeter, DSC7, PerkinElmer, Shelton, CT,

USA). The adhesives were preheated at 50℃ for 18 hours before testing. The temperature scan ranged from 25℃ to 300℃ at a heating rate of 10℃/min under nitrogen flushing (flow rate: 50 mL/min).

2.4. Thermogravimetric analysis

The adhesives preheated at 50℃ for 18 hours were subjected to thermogravimetric analysis (TGA) using a Thermogravimetric Analyzer (TGA8000, PerkinElmer). The samples were heated from 25℃ to 800℃ under nitrogen flushing at 50 mL/min and a heating rate of 10℃/min.

2.5. Evaluation of adhesive properties

The appearances of adhesives were observed. The solids content, gelation time, and pH of each adhesive were evaluated according to the JIS K6833 testing methods (JIS, 1994). For UF and MUF adhesives, about 1.5 g of adhesive was weighed prior to drying at 105 ± 1℃ for 180 ± 5 min. Then, the sample was cooled in the desiccator and weighted again. For all formulations of *Acacia mangium* BE adhesive, 1 g of adhesive was weighed and dried at 135 ± 1℃ for 60 ± 2 min. After drying, the samples were cooled in a desiccator and weighed again. The solids content of the adhesive (N) was determined as follows:

$$\text{The solids content of adhesive (\%)} = (W_d / W_s) \times 100 \quad (1)$$

where W_d and W_s are the masses of the adhesive sample after and before drying, respectively.

About 10 g of each adhesive formulation was placed in a test tube that was put in boiling water at 100℃. Adhesive was continuously stirred and the gel time was recorded as the point at which gelation occurred.

The pH of the adhesive at 23 ± 2℃ was measured with a pH meter. The viscosity (Pa s) of the adhesive at 23 ± 2℃ was measured using a Ford cup No. 4.

2.6. Rubberwood particleboard manufacturing

Single-layer particleboards were formed with the dimensions of 350 mm by 350 mm by 10 mm. The target density of the panel was 0.65 g/cm^3. Rubberwood particles were blended with each adhesive formulation contributing 10% (based on the oven-dry weight of particles) in a blender. The panels were hand-formed in a wooden block before they were subjected to hot pressing at 1.4 MPa and 150℃ for 10 mins.

2.7. Evaluation of physical and mechanical properties of particleboard

The particleboard specimens were conditioned at 20℃ and a relative humidity of 65% until a constant weight was achieved. After the samples were conditioned, their density, moisture content, thickness swelling (TS), modulus of rupture (MOR), and IB were evaluated in accordance with JIS A 5908 testing methods (JIS, 2015b).

The width, length, and thickness of each test specimen were measured, and the volume (V) was calculated. Then, the mass (m_1) of each test specimen was measured and the density was calculated:

$$\text{Density (g/cm}^3) = m_1 / V \quad (2)$$

The test specimen was put in a hot air oven at 103 ± 5℃, and when a constant mass (m_0) was achieved, it was recorded to calculate the moisture content as follows:

$$\text{Moisture content (\%)} = 100 \times (m_1 - m_0) / m_o \quad (3)$$

The TS test was conducted by soaking 50 mm by 50 mm by 10 mm samples in water at room temperature horizontally about 3 cm below the water surface for 2 h and for 24 h. The thickness at the center of the test specimen was measured before (t_1) and after (t_2) immersion in the water. The swelling in thickness after water immersion (TS) was calculated from the formula below.

$$TS\ (\%) = 100 \times (t_2 - t_1) / t_1 \quad (4)$$

The MOR test was conducted by applying a deformation at 10 mm/min rate to the surface of the test specimen. The maximum load (P) was measured, and the MOR of the individual specimen was calculated from the formula below.

$$MOR\ (N/mm^2) = 3PL / 2bt^2 \quad (5)$$

where L is the span (mm), and b and t are width (mm) and thickness (mm) of the specimen.

In the IB test, the width (b) and the length (L) of the test specimen were measured. It was adhered on a steel block, and a tension load was applied vertically to the specimen surface with a crosshead speed of 2 mm/min. The maximum loading force (P_{max}) at the time of failure was measured and the IB estimate was calculated.

$$IB\ (N/mm^2) = P_{max} / bL \quad (6)$$

2.8. Formaldehyde emissions from particleboard

The formaldehyde emissions from each type of particleboard were determined using the desiccator method in accordance with JIS A 1460 (JIS, 2015a). The amount of formaldehyde emitted was calculated based on the concentration of formaldehyde absorbed in distilled water, after placing a test specimen with a specified surface area (as close as possible to 1,800 cm^2) into a desiccator above a crystalline glass dish containing 300 mL of distilled water for 24 hours. The concentration of formaldehyde absorbed in the distilled water was determined via the Hantzsch reaction, in which the formaldehyde reacts with ammonium ions and acetylacetone to from 3,5-diacetyl-1,4-dihydrolutidine.

2.9. Statistical analyses

All results are expressed as mean ± SD. The properties of each adhesive formulation and the effects of adhesive formulation on the properties of rubberwood particleboards were evaluated using analysis of variance (ANOVA) at the 0.05 level of significance. Duncan's multiple range test was conducted to confirm significant differences between means.

3. RESULTS and DISCUSSION

3.1. Differential scanning calorimetry analysis

DSC analysis was conducted to examine the thermal behavior of the prepared adhesives, and the results are presented in Table 1. The exothermic peak represents the curing temperature of the adhesive. It was observed that some adhesive formulations exhibited a single curing peak, while others showed multiple peaks. Because the BE adhesive is composed of many chemical components, its curing peaks are broad. In contrast, the curing peaks of UF and MUF adhesives are narrow due to them consisting of only 2–3 main chemical components, such as urea, melamine, and formaldehyde. UF adhesives exhibited two curing peaks at 127℃ and 147℃, while MUF adhesive showed two curing peaks at 152.33℃ and 263.67℃. The results demonstrated that the *Acacia mangium* BE adhesive, using boric acid as a catalyst, exhibited a curing peak centered at 84.67℃ but

Table 1. Peak temperature and curing enthalpy from DSC analysis

Adhesive formulation	pH of adhesive	Peak temp (℃)	Curing enthalpy (J/g)
UF	8.45 (± 0.01)	127.67, 147	91.418, 209.287
MUF	8.99 (± 0.02)	152.33, 263.67	226.257, 104.41
BE-NaOH	10.48 (± 0.03)	95.17	431.077
BE	4.66 (± 0.02)	85	290.424
BE-Boric	3.43 (± 0.06)	84.67	222.183

The pH values are shown as mean (± SD).
DSC: differential scanning calorimetry, UF: urea formaldehyde, MUF: melamine-urea formaldehyde, BE: bark extract.

covering a broad range, between 42.52℃ and 130.91℃. It also showed a curing enthalpy (△H) of 222.183 J/g. Meanwhile, the adhesive using sodium hydroxide as a catalyst exhibited a curing peak at 95.17℃ (ranging from 55.48℃ to 135.61℃), with a curing enthalpy of 431.077 J/g. The extent of curing enthalpy is often indicative of adhesive quality, as higher values suggest more complete crosslinking that can enhance both strength and durability of the bond. The curing enthalpy (△H) of MUF adhesives was higher than that of UF adhesives (Table 1). Therefore, the bonding strength of MUF adhesive is expected to be higher than that of UF adhesive. It is expected that the BE-NaOH adhesive provides the highest bonding strength. Vázquez *et al*. (2012) found that tannin extracted from chestnut shells, when combined with paraformaldehyde as a 10% hardener (based on the tannin weight), exhibited a curing peak at 93℃ with a curing enthalpy (△H) of 3.68 J/g at pH 6. Another formulation using chestnut shell tannin, with 10% hexamine as the hardener showed curing peaks at 101.3℃ and 140.7℃, with a curing enthalpy of 30.93 J/g at pH 6. In addition, it was found that the curing enthalpy of the adhesives was influenced by the type of hardener used in the formulation. However, many factors affect particleboard properties, including mat moisture content, pressing temperature, hot-pressing pressure, and pressing time. Thus, the adhesives must be applied in the production of rubberwood particleboards to evaluate their actual strength performance and confirm this potential. Nonetheless, this study concludes that the addition of an alkaline catalyst (NaOH solution) to BE resulted in a comparatively high curing enthalpy.

3.2. Thermogravimetric analysis

TGA was carried out using one sample per adhesive formulation, due to budget limitations, and therefore SDs are not reported. Nevertheless, the obtained results provide clear comparative insights into the thermal behavior of the adhesives. The percentage of weight remaining for all adhesive formulations after being heated from 25℃ to 800℃ under a nitrogen atmosphere is presented in Table 2.

The onset temperatures of weight loss were 67.83℃, 47.42℃, and 45.87℃ for BE-NaOH, BE, and BE-Boric adhesives, respectively, indicating that the BE-NaOH adhesive exhibited the highest initial thermal resistance. At the pressing temperature of 150℃, which was employed in rubberwood particleboard production in this study, all adhesive formulations retained more than 80% of their weight (83.96%–88.36%), confirming their comparable stability under processing conditions. However, more pronounced differences were observed at elevated temperatures. At 800℃, the residues of BE-NaOH, BE, and BE-Boric adhesives were 61.48%, 25.73%, and 4.54%, respectively. These findings demonstrate that

Table 2. The temperatures and final percentage weight retained from thermogravimetric analysis of the adhesive samples

Adhesive	T_{onset} (℃)	T_{d1} (℃)	T $_{150℃}$	T_{d3} (℃)	$Residue_{800℃}$ (%)
UF	55.66* (100)	120.39 (93.48)	150 (83.96)	250.02 (71.48)	800 (9.93)
MUF	49.83 (100)	126.68 (93.63)	150 (88.36)	250.05 (72.94)	800 (0.47)
BE-NaOH	67.83 (100)	98.14 (93.08)	150 (84.56)	250.11 (78.54)	800 (61.48)
BE	47.42 (100)	75.91 (94.57)	150 (86.47)	250.04 (83.52)	800 (25.73)
BE-Boric	45.87 (100)	71.12 (95.43)	150 (87.30)	250.07 (82.95)	800 (4.54)

The values in parentheses represent the retained weight percentage of the adhesive sample during thermogravimetric analysis at the reported temperature.
* The temperature of the adhesive sample during thermogravimetric analysis, at which it retained 100% of its initial weight.
UF: urea formaldehyde, MUF: melamine-urea formaldehyde, BE: bark extract.

alkaline catalysts (NaOH) enhance thermal stability of BE based adhesives, while acidic catalysts (boric acid) reduce it. This trend is consistent with previous reports indicating that tannin incorporation into adhesive systems can improve thermal resistance, particularly during the early stages of degradation (Li *et al*., 2016b).

Acacia mangium BE is rich in condensed tannins. Polyphenolic compounds are primarily composed of flavonoid units with varying degrees of polymerization. These tannins contain phenolic hydroxyl groups that are highly reactive and responsible for polycondensation reactions essential for adhesive performance. To characterize the extract, we refer to our earlier work (Yingprasert *et al*., 2014), which reported that the BE contained 17.75 ± 2.08 mg CE/g of bark as condensed tannin content and exhibited a Stiasny number of 86.16 ± 2.63%. The Stiasny number serves as a widely accepted indicator of the reactivity of tannins toward aldehydes, especially formaldehyde, and indirectly reflects the crosslinking potential of the extract. In the present study, the adhesive formulations were prepared without aldehyde-based hardeners, but under different catalytic conditions (alkaline vs. acidic) to evaluate effects of this choice on adhesive performance. The greater curing enthalpy and bonding strength observed for the BE–NaOH formulation may be explained by classical nucleophilic aromatic substitution mechanisms that have been previously reported. Under alkaline conditions, the phenolic hydroxyl groups in condensed tannins are deprotonated to form phenolate ions, which are stronger nucleophiles. These can undergo electrophilic substitution reactions leading to the formation of methylene bridges, even in the absence of formaldehyde. This mechanism is well-established in phenol–formaldehyde and tannin–formaldehyde systems (Pizzi, 2009) and supports the enhanced thermal behavior (as observed by DSC) and bonding performance in BE–NaOH. Conversely, in the BE–Boric formulation, prepared under mildly acidic conditions, the reactivity may have been suppressed. Boric acid may form borate esters or hydrogen-bonding complexes with phenolic hydroxyl groups, which could inhibit the crosslinking reactions required for effective adhesive curing. This could explain the lower curing enthalpy for the BE–Boric formulations. These findings are consistent with prior reports indicating that the type of catalyst or hardener can significantly influence the curing kinetics and final performance of tannin-based adhesives (Anggini *et al*., 2023; Meikleham *et al*., 1994). Furthermore, earlier studies have shown that the self-condensation ability of condensed tannins can be

enhanced under alkaline conditions or by using additives like boric acid, which serve as mild crosslinking agents (Zhang *et al*., 2017). Thus, the chemical environment during adhesive formulation plays a crucial role in activating or suppressing the reactivity of condensed tannins.

3.3. Adhesive properties

The pH, viscosity, solids content, and gel time of each adhesive formulation are shown in Table 3. The commercial adhesives for particleboard production, namely UF and MUF, are alkaline with pH > 7. The UF and MUF adhesives had respective average pH values 8.45 and 8.99. The *Acacia mangium* BE adhesive was acidic with pH 4.66. In this study, boric acid and sodium hydroxide were used as catalysts to adjust the pH of the adhesive to 3 and 10, respectively. The BE-Boric and BE-NaOH adhesives had average pH values of 3.43 and 10.49, respectively. The average solids contents of UF and MUF adhesives were 67.15% and 66.78%, respectively. The average solids contents of BE adhesive (42.50%) and BE-NaOH adhesive (42.50%) were lower than those of UF and MUF adhesives. In this research, rubberwood particles were blended with each adhesive formulation contributing 10% (based on the oven-dry weight of particles) in a blender for particleboard manufacturing. The solids content of the adhesive directly affects the mat moisture content. Adhesives with a low solids content contain a comparatively large fraction of water, whereas adhesives with higher solids content contain less water. All formulations of BE based adhesives showed lower solid content compared to UF and MUF adhesives. When forming the particleboard with an adhesive formulation having a low solids content, the mat moisture content will be higher than when using an adhesive formulation with a higher solids content. This may affect the optimal control parameters during the forming process, such as pressing temperature and time, and may also influence the final properties of the particleboard. In this study, an equal weight of BE was used for the preparation of each BE adhesive. It was found that some BEs deteriorated due to heat during synthesis. The results show that NaOH did not cause a loss of BE because the solids content of BE adhesive was equal to the solids content of BE-NaOH adhesive. However, boric acid caused significant degradation of the BE during adhesive synthesis. The low solids content observed in BE-Boric adhesive (29.15%) may be related to acid-induced degradation of extractives during synthesis. Acidic catalysts, including boric acid, have been reported to promote hydrolytic cleavage of polysaccharide structures in BEs, thereby reducing the yield of solid components (Pizzi, 2023). This degradation could also contribute to the observed reduction in viscosity; however, the specific roles of hemicellulose hydrolysis

Table 3. Properties of adhesives

Adhesive formulation	pH	Viscosity (Pa·s)	Solids content (%)	Gel time (min)
UF	8.45 (0.01)	0.203 (0.004)	67.15 (0.53)	7.57 (0.02)
MUF	8.99 (0.02)	0.268 (0.003)	66.78 (0.51)	8.20 (0.63)
BE-NaOH	10.49 (0.03)	0.193 (0.004)	42.50 (0.71)	6.44 (0.02)
BE	4.66 (0.02)	0.310 (0.004)	42.50 (0.42)	29.59 (0.81)
BE-Boric	3.43 (0.06)	0.153 (0.011)	29.15 (0.64)	4.22 (0.01)

Data are presented as mean ± (SD).
UF: urea formaldehyde, MUF: melamine-urea formaldehyde, BE: bark extract.

or other chemical pathways remain uncertain. Since no direct chemical analyses were performed in this study to confirm the mechanism, this explanation should be considered as a plausible interpretation rather than a conclusive statement. Further spectroscopic and compositional studies are necessary to verify the degradation mechanisms involved. The average solids content of BE-Boric adhesive was the lowest, at 29.15%. Therefore, the mat moisture content before hot pressing may be higher than the mat moisture content when using the other adhesives. Consequently, this led to longer hot-pressing times during the manufacturing, because the excess water in the adhesive needed to be removed as steam during the hot pressing (Pizzi, 2023). In the continuous hot pressing of the manufacturing process, additives may be used to increase the solids content in the adhesive to address this problem. Modified starch mixed with polyvinyl alcohol adhesive has been used as a binder in particleboard manufacturing. Mixing polyvinyl alcohol with modified starch reduces the viscosity of modified starch and increases the solids content of modified starch (Lamaming *et al*., 2020). Solids content and viscosity are critical physical parameters for determining the processability of the adhesives in a board production line. This is an important issue that warrants further investigation.

The gel times of UF and MUF adhesives were longer than those of BE based adhesives. Catalysts are primarily employed to promote the curing kinetics of the adhesive system, resulting in shorter gelation time and improved processing efficiency. The BE adhesive without a catalyst showed the longest average gel time of 29.59 minutes. The BE-Boric adhesive gave the shortest average gel time of 4.22 minutes, while the BE-NaOH gave an average gel time of 6.44 minutes. Therefore, using the BE adhesive in rubberwood particleboard production requires the use of a catalyst to shorten the curing time of adhesive. Gel time is a measure of the reactivity of the adhesive, and the observed differences demonstrate that the additional chemicals influenced the curing behavior of the BE adhesives (Pizzi, 2023). Although the BE-Boric adhesive exhibited the shortest gel time (4.22 min), this rapid curing did not correlate with improved adhesive performance. On the contrary, its mechanical properties were the lowest among the formulations tested (MOR 5.11 N/mm^2), suggesting that accelerated gelation under acidic conditions may lead to brittle polymer networks with reduced bonding capability. Similar observations were reported by Ndiwe *et al*. (2019) for maritime pine tannin adhesives, where acid-catalyzed systems cured faster but exhibited inferior strength compared to alkaline systems. While the botanical sources differ, this comparison illustrates a general trend that highlights the trade-off between curing rate and adhesive performance. Tannin can be employed in wood adhesives when combined with silica as a hardener. Diluted water solutions of pine tannin (procyanidins) BEs, when reacted with 4% silica at pH 12 and ambient temperature, form a distinct soft gel after approximately four weeks. This outcome is not observed in the absence of silica. Specifically, catechin monomer, procyanidin, and prodelphinidin tannins form gels in the presence of dissolved silica and other weak Lewis acids due to intermolecular C2-C6/C2-C8 autocondensation. However, with natural polymeric tannins, the presence of dissolved silica inhibits intramolecular rearrangement to catechinic acid (Pizzi and Meikleham, 1995). Polyflavonoid tannins have been discovered to undergo autocondensation and hardening in the presence of small amounts of SiO_2, boric acid, and $AlCl_3$ under high-pH conditions (Meikleham *et al*., 1994).

The average viscosity of the BE−NaOH adhesive (0.193 Pa·s) was close to that of the UF adhesive (0.203 Pa·s). A possible loss of BE during adhesive synthesis may have contributed to the slight decrease in viscosity. In contrast, the addition of an acidic catalyst markedly reduced the viscosity of the BE-based adhesive. This phenomenon may be explained by the hydrolysis of

hemicellulose-derived oligomeric fragments, as previously reported by Pizzi (2023), which results in lower molecular weight fractions and, consequently, reduced viscosity. Although the present study did not investigate the detailed chemical structure or composition of the BE, it is plausible that similar hydrolytic degradation mechanisms occurred under acidic conditions. The lower viscosity may influence adhesive penetration and subsequent bonding behavior, suggesting that future research should include compositional analyses to confirm the underlying mechanisms and establish clearer correlations with adhesive performance.

3.4. Effects of adhesive formulation on physical and mechanical properties of particleboard

The physical and mechanical properties of particleboards prepared with various adhesive formulations are summarized in Table 4. The average density of the boards ranged from 0.63 to 0.71 g/cm^3. The equilibrium moisture content (EMC) at 20℃ and 65% RH varied by the adhesive system. Boards bonded with MUF adhesive had the lowest EMC (6.52%), followed by BE (7.17%), BE–Boric (7.46%), and UF (7.99%), while BE–NaOH showed the highest EMC (9.83%). Although statistical analysis (α = 0.05) indicated that the EMC of BE–Boric bonded boards was significantly lower than that of UF and BE–NaOH, this observation should not be interpreted as evidence of improved moisture resistance. The high TS and water absorption (WA) observed for BE–Boric bonded boards (2 h: 30.10% and 126.16%; 24 h: 61.69% and 144.39%) clearly demonstrate poor water resistance. According to JIS A 5908 (JIS, 2015b), the mean moisture content of Type 8 particleboard should not exceed 12% (based on oven-dry weight), and all

Table 4. Properties of rubberwood particleboards produced using alternative adhesive formulations

Adhesive formulation	Density (g/cm^3)	Moisture content (%)	TS2hr (%)	TS24hr (%)	WA2hr (%)	WA24hr (%)	IB (N/mm^2)	MOR (N/mm^2)
UF	0.69^{c}	7.99^{c}	5.31^{a}	4.88^{a}	42.33^{a}	59.41^{a}	1.39^{a}	16.59^{a}
	(0.04)	(0.47)	(2.48)	(3.07)	(14.13)	(9.29)	(0.45)	(2.73)
MUF	0.68^{bc}	6.52^{a}	4.67^{a}	9.15^{a}	40.18^{a}	58.61^{a}	1.01^{a}	18.67^{a}
	(0.03)	(0.59)	(3.21)	(4.96)	(8.81)	(5.82)	(0.49)	(5.01)
BE-NaOH	0.63^{a}	9.83^{d}	37.88^{b}	54.53^{b}	131.60^{c}	148.56^{c}	0.19^{b}	7.61^{b}
	(0.02)	(0.31)	(10.70)	(4.42)	(9.72)	(6.59)	(0.08)	(2.26)
BE	0.71^{c}	7.17^{b}	23.35^{b}	58.60^{b}	81.27^{b}	116.13^{b}	0.21^{b}	8.25^{bc}
	(0.03)	(0.18)	(4.46)	(8.39)	(10.93)	(6.83)	(0.06)	(2.23)
BE-Boric	0.64^{ab}	7.46^{b}	30.10^{b}	61.69^{b}	126.16^{c}	144.39^{c}	0.20^{b}	5.11^{c}
	(0.04)	(0.11)	(10.77)	(15.65)	(6.23)	(6.35)	(0.05)	(1.03)
JIS A 5908:2015 (Type 8)	0.40–0.90	5–13	≤ 12	≤ 12	Not specify		≥ 0.15	≥ 8

Data are expressed as averages with SDs in parentheses.
[a–d] Different letters within the same column indicate a statistically significant difference at a = 0.05.
TS: thickness swelling, WA: water absorption, IB: internal bond, MOR: modulus of rupture, UF: urea formaldehyde, MUF: melamine-urea formaldehyde, BE: bark extract.

values measured in this study fall well within the acceptable range.

The TS and WA of particleboards using BE-NaOH, BE, and BE-Boric adhesives were not significantly different from one another (a = 0.05) and were higher than those of particleboards made with UF and MUF adhesives. Water easily penetrated the particleboards bonded with *Acacia mangium* BE based adhesives, resulting in a thickness increased by 55%–62%. It was found that the weight losses of the particleboards made with BE-NaOH, BE, or BE-Boric adhesive after soaking in the water for 24 hours were 23.07%, 18.13%, and 12.78%, respectively. The percentage weight loss was higher than the percentage weight of solid adhesive sprayed on rubberwood particles. This means that the water must have dissolved components of both the rubberwood and the adhesives, even though the adhesives were hardened by curing. The laboratory results demonstrated that BE based adhesives may be suitable for interior-grade particleboard. Previous research also reported that tannin adhesive without a hardener, with a viscosity of 512 cP, solids content of 57.3%, and pH of 4.2, was used to produce particleboard (8% w/w) under pressing conditions of 160℃ and 5 MPa for 20 minutes. It was revealed that the TS after 2 and 24 hours of water immersion for the particleboards was 70.54% and 116.06%, respectively. The WA after 2 and 24 hours of water immersion for the particleboard was 109.14% and 118.62%, respectively (Nath *et al*., 2018). In Thailand's wood-based panel industry, it is common practice to spray wax onto rubberwood flakes simultaneously with commercial adhesives such as urea-formaldehyde or melamine-urea-formaldehyde resins during the manufacturing of particleboard. This combined application aims to enhance the dimensional stability of the panels, particularly by improving TS and WA performance. Xu *et al*. (2009) demonstrated that the incorporation of wax substantially lowered TS and WA after 24 hours of water immersion, when compared to untreated bagasse particleboard. Investigating the effects of wax incorporation into BE based adhesives represents a promising direction for future research aimed at enhancing TS and WA properties of rubberwood particleboard. Various other strategies have also been employed to enhance particleboard's dimensional stability. For example, Wanishdilokratn and Wanishdilokratn (2024) reported that incorporating silicon dioxide into particleboards made from teak sawdust resulted in significant reductions in both WA and TS after water immersion. Additionally, Aisyadea *et al*. (2023) found that increasing the proportion of fine particles and the resin content in the board formulation contributed to improved resistance to water uptake and swelling. One significant limitation of BE based adhesives is their poor water resistance, which restricts their broader application in the wood-based panel industry. Overcoming this limitation could greatly expand their usability. In this context, Zhang *et al.* (2019) reported that the water resistance of tannin-based adhesives can be effectively improved through cross-linking with furfuryl alcohol, glyoxal, and epoxy resin under acidic conditions. The addition of Tinnevelly senna seed flour to a natural adhesive, composed of condensed tannin from *Acacia mimosa* and hexamethylenetetramine at pH 10, significantly reduced TS and WA of particleboards (Eghtedarnejad and Mansouri, 2018). Incorporating a small amount of potato starch into the particleboard formulation containing kenaf fiber was found to enhance mechanical strength, IB, and contact angle, while simultaneously reducing WA and TS. The bonding mechanism in the particleboard can be attributed to the formation of intermolecular hydrogen bonds between the amylopectin and amylose components of potato starch and the cellulose, hemicellulose, and lignin in kenaf particles (Nadhari *et al*., 2024).

The IB strength of particleboards produced with UF and MUF adhesives was significantly higher than that of boards bonded with BE-NaOH, BE, and BE-Boric adhesives. However, no statistically significant differences in

IB were observed among BE-NaOH, BE, and BE-Boric formulations (α = 0.05). Notably, all IB values obtained in this study met the minimum requirement of 0.15 N/mm^2 specified in JIS A 5908 (Type 8; JIS, 2015b). These results indicate that both alkaline and acidic conditions during adhesive synthesis had comparable effects on the IB of rubberwood particleboards bonded with BE based adhesives.

Previous studies reported that boric acid enhances the autocondensation reaction of tannins during the manufacture of poplar plywood, reducing both curing temperature and time while simultaneously increasing the adhesive modulus of elasticity (Efhamisisi *et al*., 2016). Moreover, boric acid was shown to improve bonding performance and provide resistance to fungal degradation. The *Acacia mangium* tannin, treated with a NaOH/urea solution, underwent depolymerization, resulting in a low-molecular-weight tannin solution. The main breaks in condensed tannin occurred at C-4 and C-8 positions, and several ethers were detected. These bond cleavages facilitated the uniform distribution of tannins in the aqueous solution, thereby enhancing the bond strength of the modified tannin adhesive (Liu *et al*., 2020).

The MOR of particleboards bonded with UF (16.59 N/mm^2) and MUF (18.67 N/mm^2) adhesives was higher than that of particleboards prepared with BE adhesives. No significant differences in MOR were observed between BE-NaOH and BE, or between BE and BE-Boric adhesives (α = 0.05). BE-NaOH and BE adhesives yielded MOR values of 7.16 N/mm^2 and 8.25 N/mm^2, respectively, while BE-Boric showed a lower MOR of 5.11 N/mm^2. Although below those for UF and MUF adhesives, these results are consistent with Nath *et al*. (2018), who reported MOR values of tannin-based particleboards comparable to those bonded with UF adhesives. All particleboards bonded with *Acacia mangium* BE adhesives in this study nonetheless met the Type 8 requirements specified in JIS A 5908 (JIS, 2015b). The comparatively lower mechanical performance of BE adhesives can be attributed to the absence of hardeners or crosslinkers, which play a critical role in establishing a robust three-dimensional network. Although UF and MUF adhesives were also used without hardeners in this study, their inherent high tack strength contributed to stronger bonding during mat formation. By contrast, BE adhesives exhibited low tack strength, as observed during manual board forming from wood particles sprayed with these adhesives that appeared relatively dry, with limited adhesive retention on particle surfaces.

Viscosity and solids content measurements further support these observations. BE-NaOH and BE-Boric adhesives displayed lower viscosity than commercial UF and MUF adhesives, resulting in thinner adhesive films upon spraying. Due to the high water content, BE adhesives penetrated into the wood substrate rather than remaining on the surface, reducing effective glue-line coverage and consequently lowering bonding strength. In contrast, UF and MUF adhesives typically incorporate starch to enhance viscosity, solids content, and tack strength. The lower values of these properties in BE adhesives explain their reduced mechanical performance.

Improving the solids content, viscosity, and tack strength of BE adhesives is therefore essential for enhancing their industrial applicability. It was difficult to directly evaluate the cold tack strength of the adhesive during particleboard formation. However, examining the adhesive coverage thickness on wood particle surfaces using microscopy may provide quantitative data to better describe the cold tack behavior of the adhesive. Incorporation of starch-based components has been shown to improve adhesive performance by crosslinking with organic acids. For example, Muhaimin *et al*. (2025) demonstrated that starch–citric acid adhesives form ester bonds, thereby increasing crosslinking density and improving bonding properties in bamboo particleboards. Citric acid also enhances starch dispersion, resulting in

more uniform adhesive coverage. Similarly, boric acid has been explored as a potential crosslinker to improve durability and biodegradation resistance, although in the current study it did not enhance board properties. Nevertheless, its role in improving dimensional stability and biological resistance remains promising for future applications.

Other strategies may include incorporating suitable fillers to increase adhesive solids content and improve cold tack, an important property influencing mat integrity prior to hot pressing. Inadequate cold tack can cause deformation of pre-pressed mats, thereby reducing pressing efficiency. Additives such as polyvinyl acetate emulsion have been shown to dramatically increase cold tack in soy protein-based adhesives (Ye *et al*., 2024). Similarly, potato starch has been reported to enhance the MOR of kenaf-based particleboards by forming dextrins during hot pressing, which provide effective tack and adhesion (Nadhari *et al*., 2024).

In addition, previous studies have demonstrated that blending tannins with hardeners or co-binders (e.g., hexamine, corn flour, or seed flours) significantly improves mechanical properties (Eghtedarnejad and Mansouri, 2018; Moubarik *et al*., 2013; Santos *et al*., 2017). These findings suggest that the use of suitable hardeners and fillers could similarly enhance the performance of BE adhesives. Furthermore, the incorporation of hydrophobic agents, such as wax, may improve water resistance and dimensional stability of the final product.

Overall, the findings of this study indicate that while *Acacia mangium* BE adhesives provide environmentally friendly and formaldehyde-free alternatives, their performance remains inferior to UF and MUF adhesives. Future work should therefore focus on (i) incorporating starch-based components or fillers to increase solids content, viscosity, and tack strength, (ii) identifying effective crosslinkers and hardeners to promote curing and bonding strength, and (iii) evaluating the role of water-repellent agents to enhance durability. These approaches hold potential for advancing BE-based adhesives toward industrial application in particleboard manufacturing.

3.5. Formaldehyde emissions from particleboard

The formaldehyde emissions of the particleboard panels produced with the various adhesive formulations are summarized in Tables 5. The results show that the formaldehyde emissions of rubberwood particleboard produced using UF and MUF adhesives were 5.30 mg/L and 2.58 mg/L, respectively. According to the JIS A 5908 standard (JIS, 2015b), the maximum permissible level for the F** rating is 1.5 mg/L. Therefore, both UF and MUF adhesives exceeded this limit. In contrast, the emissions from particleboards bonded with BE-NaOH, BE, and BE-Boric adhesives were 0.013, 0.058, and 0.047 mg/L, respectively, all of which complied with the F**** requirement. These extremely low emission levels provide clear health safety benefits and market competitiveness for interior-grade particleboard in Thailand's rubberwood industry. Additionally, the low emissions

Table 5. The formaldehyde emissions from rubberwood particleboard formed with each adhesive formulation

Adhesive	Formaldehyde emissions (mg/L)	Classification
UF	5.30 (0.10)	Failing F**
MUF	2.58 (0.01)	Failing F**
BE-NaOH	0.013 (0.001)	F****
BE	0.058 (0.001)	F****
BE-Boric	0.047 (0.001)	F****

Data are presented as mean ± (SD with n = 3).
Data from JIS (2015b).
UF: urea formaldehyde, MUF: melamine-urea formaldehyde, BE: bark extract.

indicate potential cost-effectiveness in markets with strict formaldehyde regulations.

It should be noted that the UF and MUF adhesives used in this study were commercial products with undisclosed formaldehyde-to-urea and formaldehyde-to-melamine molar ratios. Since such ratios strongly influence formaldehyde emission levels, this represents a limitation when making direct comparisons with BE adhesives. For comparison, Santos *et al*. (2017) reported that particleboards bonded with chestnut shell tannin extracts and hardeners such as tris(hydroxymethyl)nitromethane, glyoxal, and hexamethylenetetramine achieved emissions within the E0 commercial standard. While their study demonstrated the feasibility of tannin-based adhesives in producing low-emission particleboards, our study differs in several key aspects: (1) the use of locally abundant and low-cost *Acacia mangium* BE, (2) preparation of adhesives without aldehyde-based hardeners, and (3) systematic evaluation under different pH conditions using both alkaline (NaOH) and acidic (boric acid) catalysts. These factors not only maintain extremely low formaldehyde emissions but also provide insights into the influence of pH on adhesive reactivity and crosslinking, which was not addressed in Santos *et al*. (2017). Furthermore, the present study demonstrates that *Acacia mangium* BE adhesives can satisfy mechanical and emission requirements simultaneously at the laboratory scale, highlighting the potential of a novel, sustainable, and economically feasible alternative to commercial UF/MUF adhesives for interior-grade particleboard.

4. CONCLUSIONS

This study confirmed the feasibility of using *Acacia mangium* BE as a bio-based, formaldehyde-free adhesive for rubberwood particleboard production. Adhesives were prepared without aldehyde-based hardeners and were catalyzed under different pH conditions to assess their performance. Sodium hydroxide (alkaline) and boric acid (acidic) catalysts significantly influenced adhesive behavior. Among the tested formulations, BE–NaOH exhibited the highest curing enthalpy and thermal stability, indicating greater reactivity and crosslinking potential under alkaline conditions. Rubberwood particleboards bonded with BE–NaOH, BE, and BE–Boric adhesives met the minimum IB and MOR requirements for Type 8 particleboard specified in JIS A 5908 (JIS, 2015b), although their IB values were close to the lower threshold and their overall mechanical performance remained poorer than those of particleboards bonded with commercial UF and MUF adhesives. In addition, the high TS and WA with BE adhesives indicated limited water resistance. The molar ratios of commercial UF/MUF adhesives were not available, representing a limitation for direct mechanistic comparison.

Despite these constraints, all particleboards bonded with BE-based adhesives achieved F**** formaldehyde emission ratings, demonstrating a clear advantage in terms of health safety and environmental impact, and indicating potential applicability in Thailand's rubberwood particleboard industry.

To improve industrial applicability, future studies should focus on formulation optimization. Incorporation of starch-based components or suitable crosslinkers (e.g., citric acid, boric acid) may enhance crosslinking density, bond strength, and curing efficiency. The use of natural fillers can increase solids content and improve cold tack, while water-repellent agents, such as wax, can enhance dimensional stability and water resistance. Selection of appropriate hardeners may further accelerate curing and strengthen adhesive bonds. These strategies specifically address the identified limitations in water resistance (TS and WA) and bonding performance, providing a pathway for advancing BE adhesives as sustainable, formaldehyde-free alternatives to synthetic resins for wood-based panels.

CONFLICT of INTEREST

No potential conflict of interest relevant to this article was reported.

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