# Bonding Characteristic of Gambir Tannin-based Adhesive on Tusam Wood (*Pinus merkusii*) in Various Ages: Effects of Gambir Leaves Condition and Extender Addition

Adi Santoso, Erlina Nurul Aini, and Dina Alva Prastiwi

**Abstract**

Exploration of bio-based wood adhesive as an alternative to conventional adhesives such as urea-formaldehyde (UF) and phenol-formaldehyde (PF) is an interesting topic to be investigated further due to the increase in human awareness of environmental sustainability and health. Among several bio-based adhesives, the tannin-based adhesive has considerable potential to be developed as a commercial wood adhesive. In Indonesia, one of the materials that have high potential as a raw material for making tannin-based adhesives is gambir leaves. In this study, the effects of leaf freshness, extender content, and wood age on the quality of the laminated product were studied. The results showed that all three factors significantly affect the compressive shear strength of the laminated wood. The condition of fresh leaves, 0% extender content and 27 years old wood produced a laminated product with the highest compressive shear strength i.e., 109.30 kg/cm². All laminated wood products in this study meet the compressive shear strength requirement of the Indonesia National Standard (SNI). Moreover, all laminated wood glued using tannin adhesive from fresh leaves without the addition of an extender are also able to meet the Japanese Agricultural Standard (JAS) requirement of compressive strength. The research results also show that gambir tannin adhesive has properties and bonding strength that compete with PF adhesive. From these results, gambir tannin-based adhesive demonstrates an ability to serve as an alternative to conventional adhesive commonly used in the laminated wood industry.

**Keywords:** Compressive shear strength, extender content, gambir tannin-based adhesive, leaf freshness, tusam wood.

## Introduction

The rising awareness of both environmental sustainability and human health has triggered the efforts to use renewable and safe materials for health in various aspects of life. Regarding wood adhesive, this was done by utilizing biomaterials to produce adhesive that can serve as an alternative to the conventional adhesives generally used in the wood processing industry. Conventional adhesives such as urea-formaldehyde (UF) and phenol-formaldehyde (PF) have advantages, i.e., they are relatively inexpensive and have excellent performance as adhesives. The excellent adhesive performance of UF and PF was shown by the strong and stable characteristics of the product bonded by both adhesives (Mamza *et al*. 2014). However, UF and PF are adhesives that are derived from non-renewable materials and produce formaldehyde emissions that are harmful to human health (Raydan *et al*. 2021).

Some researchers found that renewable and non-toxic materials like starch (Gadhave *et al*. 2017), lignin (Dongre *et al.* 2015), and tannin (Dhawale *et al*. 2022) have considerable potential to be developed as a wood adhesive. Their potential as wood adhesives was related to their massive availability in nature, which led to their economical price compared to conventional adhesives. Natural based adhesive also has a bonding ability that can be on par with conventional adhesives (Müller *et al*. 2007; Gonultas 2018). Among the natural material-based adhesives, tannin adhesive is quite an attractive option for further development, especially since the development of tannin adhesive has reached the commercialization stage (Pizzi 2016). Tannin comprises several complex organic compounds and has high reactivity toward aldehydes and other reagents, especially in condensed forms (Pizzi, 2008; Zhang *et al*. 2022). The high reactivity was attributed to high phenolic compound content in tannin, which makes tannin can be used as an adhesive or as a substitute for phenols in adhesives (Pizzi 2006; Zhou and Du 2019). According to Santoso *et al.* (2012), tannin-formaldehyde adhesive emits significantly less formaldehyde than UF and PF adhesives, which becomes an additional advantage of tannin-based adhesive.

As a country with high biodiversity, Indonesia has many sources of tannins that can be used to produce adhesives. Tannin from gambir (*Uncaria gambir* (Hunter) Roxb.) leaf is one of the many sources of tannins available in Indonesia. Gambir plants in Indonesia are mainly present on Sumatra Island, especially in West Sumatra Province, where about 80% of total national gambir production comes from this province (Isnawati *et al*. 2012). In 2020, BPS (2021) recorded that the gambir plantation area in West Sumatra was about 28.016 ha. The gambir plantation area is predicted to increase further in West Sumatra and other potential locations in Indonesia. It is due to the Ministry of Agriculture's plan to increase Indonesia's gambir production up to 500% between 2020-2024 (Kementan 2020).

Adhesive from gambir extract has also been shown to have good bonding performance that can meet quality standards from Indonesia and Japan. Malrianti *et al*. (2018) did research to produce a cold-press adhesive from gambir-tannin extract with an addition of 10% hexamethylenetetramine. That research found that laminated wood bonded using gambir-tannin cold setting adhesive with a glue spread rate of 200 g/m² and pressing time of 24 h has compressive shear strength between 11.5 to 21.1 kg/cm². Other research about gambir-tannin as an adhesive was also done by Sucipto *et al*. (2020). The application of gambir-tannin-sucrose as a hot-press adhesive in the production of bamboo particleboard resulted in excellent internal bonding strength of the board that reached 0.89 MPa (Sucipto *et al*. 2020). The particleboard manufacturing condition used was gambir-tannin/sucrose composition of 25/75wt% and press temperature of 200°C for 10 minutes. The effective polymerization reaction via the methylene bridge occurred between tannin-hexamethylenetetramine and tannin-hydroxy-methyl-furfural (5-HMF) derived from sucrose contributed to the excellent bonding strength of these gambir adhesives.

The glued product quality is broadly influenced by the adhesive material, adherend, the gluing process, and the product use conditions (Sucipto and Ruhendi 2012). Regarding the adhesive factor, an adhesive component consisting of the main component and additional components, such as an extender, plays a crucial role in determining the quality of the adhesive and the bonded product. In bioadhesive that used biomass extract as the main component, the condition of the extracted raw material, like its freshness, will affect the quality of the produced extract (Abdul Razak *et al*. 2014). While in adherend factors such as wood, the natural properties of wood will also have a major influence on the bonding quality and the overall glued product properties. According to Basri *et al*. (2012), the natural properties of wood can be influenced by its age. The difference in natural wood properties may affect its adhesion properties. Therefore, this study investigated the effect of the freshness condition of gambir leaves as a source of tannins on the quality of the gambir tannin-based adhesive. This study also examines the effect of leaf freshness, extender content, and wood age on the laminated product's compressive shear strength. This research was expected to provide information for the benefit of further development of gambir tannin-based wood adhesive. Tusam wood (*Pinus merkusii* Jung et de Vriese) is used as the substrate because it is one of the most accessible types of wood to find in Indonesia. It also has been widely used in wood composite products manufacture.

## Materials and Methods

The materials used in this study were gambir (*Uncaria gambir* (Hunter) Roxb.) leaves in fresh and dry conditions, tusam wood (*Pinus merkusii* Jung et de Vriese), distilled water, technical grade formaldehyde solution (HCOH, 37%), and technical grade sodium hydroxide solution(NaOH, 40%). An industrial wheat flour also used in this research, as an extender. Fresh gambir leaves were obtained by taking them directly from the tree during the gambir harvest period, while dry gambir leaves were taken from leaves that fell to the ground. The tusam wood used in this study was in age 17, 21, 23, 27, and 28 years old. The equipment needed in this research was a measuring cup, a measuring flask, a burette, a soxhlet apparatus, a porcelain dish, a water bath (Laboratory Water Bath WNB 29 Ring, Memmert, Germany), a cold press (Local Build, Indonesia), an oven (Universal Drying Oven UN 500, Memmert, Germany), a moisturemeter (Mini-Ligno E/D Moisturemeter, Lignomat, USA) a pH meter (pHTestr 30, Eutech Instruments, Singapore), a universal testing machine (LLYOYD Instrument EZ 20 Material Testing Machine, Ametek Inc., USA), a pycnometer (PICNO-25 ml, IWAKI, Indonesia), a viscometer (Viscotester VT-04E, Rion, Japan), an extractor (Local Build, Indonesia), and a reactor (Local Build, Indonesia).

### Analysis of Gambir Leaves Characteristics

The analysis of gambir leaf characteristics was carried out in this study by water content and tannin content evaluation. Both analyses were carried out with three (3) replications per type of leaf condition. The water content of gambir leaves was determined using the oven drying method with a temperature of ±105°C following the AOAC method (2005). Meanwhile, the tannin content determination was completed using the permanganometry method (Depkes RI 1995). The analysis was done by dissolving gambir leaves into distilled water using a heating process. A number of the filtered solutions were mixed with the indigo carmine indicator. Then the solution mixture was titrated using 0.1 N Potassium Permanganate (KMNO4). Titration was finished until the solution color changed from blue to green and finally golden yellow. The volume of KMNO4 added was recorded. Blank titration was also carried out using KMNO4 solution to calculate tannin content. The amount of tannin content contained in gambir leaves was calculated using the equation from Atanassova and Christova (2009) as stated in Formula 1.

$$KT = ((V1-V2) \times 0{,}004157 \times fp)/B \times 100 \quad \text{(Formula 1)}$$

with:
KT = Tannin content (%)
V1 = KMNO4 volume for tannin titration (ml)
V2 = KMNO4 volume for blank titration (ml)
fp = dilution factor
B = gambir leaf sample weight (g)
0,004157 is the equivalence number of tannins to 1 ml KMNO4 0.1 N

### Tannin Extraction from Gambir Leaves

The initial stage of the gambir leaves tannin extraction process is done by soaking the gambir leaves in an extractor using distilled water heated to a temperature of 70-80°C. This gambir extraction method followed the extraction method done in Santoso Abdurrahman (2016) and Hendrik *et al*. (2019). Leaf composition : distilled water used was 1 : 3. After that, the mixture of gambir leaves and distilled water was heated for about 1 hour at a temperature of ±90°C. The gambir leaves and distilled water mixture were stirred every 15 minutes during the heating or boiling process. After the heating process was complete, the mixture of gambir leaves and distilled water was cooled and then filtered using general filter paper. After that, the gambir leaf residue from the first extraction was extracted again in the same way for two (2) times more. The total extraction cycles performed were three (3) extraction cycles. The obtained liquid tannin was then stored in a closed container.

**Preparation of Gambir-Tannin-Formaldehyde Adhesive**

The manufacture of tannin-formaldehyde (TF) adhesive was carried out in several stages. The first step was to mix the liquid gambir-tannin extract with 37% formaldehyde solution and 40% NaOH in the reactor. The ratio of tannin extract : 37% formaldehyde : 40% NaOH used was 30 : 6 : 1. The pH of the solution was adjusted to 11 using NaOH (40%). The mixture was then heated to 90°C for 3 hours. During this, it was hypothesized that a polymerization reaction between tannin and formaldehyde via methylene bridge linkage would occur, resulting in a tannin-formaldehyde polymer (Zhou and Du 2019). The solution was agitated at a slow speed, and the pH was controlled at 11 every 1 hour using NaOH. After heating at 90°C for 3 hours, the reactor temperature was increased to 100°C, and let the reactor was run for ±1 hour to reduce further the water contained in the solution. If the water content was further reduced, it was expected that the TF adhesive's viscosity would increase and could meet the viscosity standard of PF adhesive. The reactor was then turned off. The solution in the reactor was conditioned until it reached room temperature and a pH of 11. Next, the TF solution was stored in a closed container. Prior to applying TF adhesive, wheat flour extender was added at 0%, 5%, 10%, 15%, and 20% levels based on the weight of the adhesive. The mixture was then stirred manually until homogenous.

**Analysis of Gambir-Tannin-Formaldehyde Adhesive characteristics**

The adhesive characteristics were analyzed, including visual tests, gelation time, solid resin content, viscosity, acidity (pH), and specific gravity. Visual testing was done by observing the color and the presence of impurities in the adhesive. Gelation time evaluation was completed by boiling the TF adhesive at ±100°C, which had been put inside a test tube before until it showed a limited flowing characteristic when the test tube containing TF adhesive was tilted. The assessment of solid content was carried out using the gravimetric method. TF adhesive sample was dried in the oven at 103±2°C until it reached constant weight. Adhesive weight before and after the drying process was used to calculate its solid content. Viscocity, acidity, and specific gravity properties were evaluated at room temperature conditions using a viscometer, pH meter, and pycnometer, respectively. All adhesive analyses were carried out based on the standard SNI 06-4567-1998 (SNI 1998) with three (3) replications for each treatment.

**Laminated Wood Manufacture**

The manufacture of laminated products from tusam wood started with wood preparation. At this stage, the logs were cut into 20 cm x 6 cm x 1 cm. After being cut, the wooden slats were then dried using an oven at a temperature of ±50°C to reach a moisture content of ±12%. The next stage was the gluing process using TF adhesive. The extender level used is 0%, 5%, 10% 15%, and 20% based on adhesive weight. The adhesive was spread evenly onto the surface of the wooden slats. The glue spread rate used is 170 g/m². After the adhesive was spread evenly, the wooden slats were cold-pressed at room temperature for 24 hours. The number of replications used in the manufacture of laminated wood was three (3) replications per treatment combination.

**Evaluation of Laminated Wood Compressive Shear Strength**

The laminated wood was left for ±7 days for conditioning before the product property was evaluated. The evaluation of laminate product property was only in the form of compressive shear or block shear strength testing. The shear strength analysis of the sample in this study was carried out based on the JPIC No. standard. 243 (2003) using a Universal Testing Machine.

**Analysis of Tusam Wood Extractive Content**

Analysis of wood extractive content completed in this research included the determination of cold-water-soluble extractive, hot-water soluble extractive, and alcohol-benzene soluble extractive contents. Analysis of cold-water and hot-water soluble extractive content was done by referring to the method in SNI 01-1305-1989 (SNI 1989a). Meanwhile, the alcohol-benzene soluble extractive content was tested based on SNI 14-1032-1989 (SNI 1989b). All measurements of extractive content were repeated 3 times for each wood age. For cold-water extractive content analysis, it was done using several steps. Firstly, the wood sample was immersed in room temperature water for 48 h, then dried the wood sample using the oven for 4 hours at 103±2°C. The drying process was done until the wood sample weight was constant. Hot-water extractive content analysis was conducted by putting an Erlenmeyer flask with a mixture of wood samples and distilled water into a water bath containing boiling water for 3 hours. The hot-water extracted wood sample was then dried using the same method as drying the wood sample after cold-water extraction. The cold-water and hot-water extractive content were calculated using their weight before and after the extraction.

Alcohol-benzene extractive content evaluation was completed by putting the wood sample in filtering paper (Whatman No. 1 filter paper). The filter paper was put into a petri dish and then placed inside a soxhlet apparatus. A 1:2 alcohol-benzene solution was added inside the apparatus. The extraction process was conducted using a water bath for 6 hours. After that, the filtering paper containing samples was removed from the apparatus. The produced extraction solution was steamed until nearly dry. The residual evaporation was then heated in the oven at 103±2°C for 3 hours or until it reached constant weight. The alcohol-benzene extractive content was estimated using wood sample dry weight before extraction and residual evaporation dry weight.

**Statistical Analysis**

All data were recorded to calculate its average value and standard deviation using the Microsoft Excel program (Microsoft Office Excel 2013, Microsoft, USA). For the extractive content analysis data and compressive shear strength data, the 3-way analysis of variance using the IBM SPSS program (IBM SPSS Statistics Base 22.0 for Windows, IBM, USA) was also carried out, continued by Tukey's HSD test to understand subgroup differences among the different experimental and control groups (Lee and Lee 2018). Tukey's HSD test was chosen for this research data analysis because it tests all pairwise differences. It is also simple to compute and can reduce the probability of making a Type I error (McHugh 2011). Type I error in statistics means rejecting the null hypothesis when it is actually true; hence committing this type of error is considered riskier compared to other errors (Kim 2015).

Table 1. Water content and tannin content of gambir leaves

| Leaf condition | Water content (%) | Tannin content (%) |
|---|---|---|
| Fresh | 63.67±0.86 | 18.16±0.88 |
| Dry | 17.16±0.41 | 15.91±0.94 |

Table 2. Characteristics of tannin formaldehyde adhesive from gambir leaves

| Adhesive Characteristics | Adhesive type | | |
|---|---|---|---|
| | TGFB | TGFK | PF standard |
| Color | Brown-black | Brown-black | Brown-black |
| Odor | Phenolic typical | Phenolic typical | Phenolic typical |
| Gelation time (minute) | 94 | 87 | 30-60 |
| Solid resin content (%) | 26.01 | 28.95 | 41-43 |
| Viscosity (25±1°C), (Poise) | 2.85 | 2.95 | 1.5-3 |
| Acidity / pH | 10.35 | 10.32 | 10-13.6 |
| Specific gravity | 1.08 | 1.06 | 1.18-1.20 |

Note : TGFB = Tannin formaldehyde adhesive from fresh gambir leaf extract
TGFK = Tannin formaldehyde adhesive from dry gambir leaf extract
PF = Phenol formaldehyde
*PF standard refers to the properties of PF adhesive stated in SNI 06-4567-1998 (SNI 1998)*
Gelation time was measured at a temperature of 100°C

## Results and Discussion

### Gambir Leaf Characteristics

As the main component of tannin-based adhesives, the quality of tannins was quite an important parameter related to their quality as a wood adhesive. The tannin quality, such as tannin content, can be affected by the freshness condition of the tannin source material (Kanto *et al*. 2008). The analysis results of the gambir leaves used as tannin sources in the manufacture of tannin-based adhesives in this study are listed in Table 1.

From the results in Table 1, it can be seen that the tannin yield of fresh gambir leaves was about 3% higher than dry gambir leaves. Similar results were obtained in Stewart *et al.* (2000) study. The extraction from *Calliandra calothyrsus* leaves without drying treatment produced higher tannin content than leaves with drying treatment. This could be explained by the occurrence of thermal degradation of phenolic compounds, including tannins, due to the heat present during the drying process (Jeong *et al*. 2004). The degradation of phenolic components does not only occur at high drying temperatures but also occurs at air drying temperatures (Miranda *et al*. 2010). In addition, Martin-Cabrejas *et al.* (2009) theorized that the decrease in tannin content in dried gambir leaves was due to changes in the polyphenol structure during the drying process. The change in tannin structure makes tannin extraction more difficult. Based on the tannin content in the production of gambir tannin-based adhesive, it would be better to use fresh gambir leaves as a tannin source. However, more caution in storing fresh gambir leaves was needed to avoid leaf decomposition. Fresh gambir leaves have a moisture content of 64%, which is much higher than dry gambir leaves, i.e., 17%. Leaf water content in the range of 60% is the optimal water content value to support the leaf decomposition process (Kurnia *et al*. 2017). The decomposition process that occurs in biomaterials will cause a change in their chemical content, such as the reduction of polyphenols content, including tannins (Zhang *et al*. 2013). Therefore, in storing fresh gambir leaves for tannin production, extra care needs to be taken so that the quantity and quality of the produced tannins do not decrease. Fresh gambir leaves, in particular, should not be stored in direct contact with soil to minimize decomposition.

**Gambir Tannin Formaldehyde Adhesive Characteristics**

The result of the Gambir-tannin-formaldehyde (TF) adhesive characteristics evaluation is in Table 2. The results showed that all adhesive characteristics of TGFB and TGFK adhesives except solid resin content, gelation time, and specific gravity met the standard for PF adhesive according to SNI 06 4567 (1998). The viscosity of the adhesive will affect its flow and even the distribution level of the adhesive on the bonded material surface (Karliati *et al*. 2014). Meanwhile, the adhesive pH in this study was deliberately conditioned under alkaline pH conditions. Alkaline pH condition was aimed to slow down the adhesive polymerization reaction during storage, hence making the adhesive remain stable in liquid form (Santoso 2003). Besides maintaining the stability of the liquid adhesive form, an alkaline pH of adhesive also resulted in a more durable adhesion or bonding than the adhesive with an acid pH. (Wangaard *et al*. 1946; Blomquist *et al*. 1949).

The specific gravity of a liquid product, like adhesive, can reflect its relative weight and molecular weight (Stauffer *et al*. 2008). In this research, gambir TF adhesive's specific gravity was slightly lower than PF adhesive. This could be indicated that gambir TF adhesive polymer molecular weight was also lower than PF adhesive. Molecular weight and the degree of polymerization have a close relation. The high degree of polymerization tends to result in a high polymer molecular weight (Balani *et al*. 2014; Saminathan *et al*. 2014). Makkar *et al*. (1990) and Zanetti *et al*. (2014) stated that high tannin content might lead to high polymerization. As mentioned previously, gambir tannin content obtained in this research was considered low (16-18%). This could lead to the low degree polymerization and specific gravity of gambir TF adhesive compared to PF adhesive.

The gelation time test showed that the gelation time of TGFB and TGFK adhesives were about 56% and 45% longer than the upper limit value at the standard gelation time of PF (60 minutes). Gelation time is the time required for an adhesive to form a hard gel or have a very high viscosity, which makes it can not be used anymore (Aulitata *et al*. 2021). Dunky and Pizzi (2002) stated that the reactivity of an adhesive could be seen from its gelation time. The low reactivity of an adhesive might lead to a long gelation time. It can be concluded that the gambir TF adhesive in this research has a longer gelation time due to its lower reactivity compared to the PF adhesive. The low tannin content in gambir extract prompted this lower reactivity of gambir TF adhesive. Hafiz *et al*. (2020) found that with the increase of tannin addition on tannin-phenol-formaldehyde adhesive, its gelation time was increased too.

Besides its content, the gambir tannin's low reactivity could have a role too in the long gelation time of the gambir-TF adhesive. In addition to its composition, the low reactivity of gambir tannin may contribute to the long gelation time of the gambir-TF adhesive. Khiari *et al.* (2017) conducted research using the lyophilization technique to increase tannin reactivity. The degree of reactivity of lyophilized tannin was found to be even greater than that of condensed tannin, resulting in a shorter gelation time for lypholized-tannin adhesive compared to condensed tannin-adhesive. Kassim *et al.* (2011) discovered that the stiansy number of gambir-tannin extracted with hot water was 12 to 28% lower than the stiansy number of ethanol-extracted tannin and ethyl ethylene-extracted tannin. The stiansy number could be used to predict the amount of condensed tannin, a highly reactive tannin component, in an extract (Garro Gavlez *et al.* 1996). Tannins with a lower stiansy value tended to be less reactive. The hot-water extraction technique utilized in this study may not be ideal for extracting condensed tannin. Consequently, the reactivity of gambir tannin was likely low; consequently, the gelation time of TF adhesive was prolonged.

In contrast to the gelation time value, the solid resin content value of TGFB and TGFK adhesives was 14-17% lower than the solid resin content in the PF standard (43%). Solid resin content describes the amount of resin formed as a result of the polymerization of TF adhesive. One of the reasons that caused the low value of gambir TF adhesive's solid content was the low value of tannin content. The low tannin extract was predicted to make the reaction between tannin and formaldehyde to form TF or resin polymer less effective, causing the lesser solid content. Besides that, the low solid content was probably also caused by the high content of non-tannin components in this research's gambir extract, which was about 80%. Pizzi and Scharfetter (1978) explained that non-tannin components would reduce the actual solid content of TF adhesive because they did not take part in the reaction to form resin. Therefore, they could hamper the final properties of the obtained resin.

The solid resin content of the adhesive can affect other properties of the adhesive, such as bonding ability (Xing *et al*. 2006; Chen *et al*. 2015). Bonding ability has a linear relationship with solid resin content. The higher the solid resin content, the higher the number of molecules in the polymer expected to play a role in the reaction between adhesive and adherend. This opinion was also conveyed by Sucipto *et al*. (2020), who stated that the bonding power of an adhesive could be represented by its solid resin content. Adhesive with high solid resin content typically also has high bonding ability due to the high content of the main active binder in the adhesive able to react with wood. With a lower solid resin content, it became a concern that gambir TF adhesive in this study will have lower bonding strength than the standard PF adhesive.

Based on the result of adhesive characteristics evaluation, it can be seen that the reactivity of gambir-TF adhesive had not reached the optimum level yet. According to Griyanitasari *et al*. (2019), DSC analysis results of the gambir extract showed that the melting point of tannin gambir occurred at ±200°C. This melting point could represent the optimum temperature for gambir-tannin to react with another compound. The research done by Sucipto *et al*. (2020) also supports this statement. In Sucipto *et al*. (2020) research, the insoluble matter of gambir adhesive reached the highest value when the adhesive was heated at a temperature of around 200°C. The measurement of insoluble matter content could indicate the degree of polymerization of the adhesive (Umemura *et al*. 2017). In order to produce TF adhesive with optimal properties, it may be necessary to use a higher temperature or to add other compounds that can reduce the optimal tannin-gambir reaction temperature during the production of gambir TF adhesive.

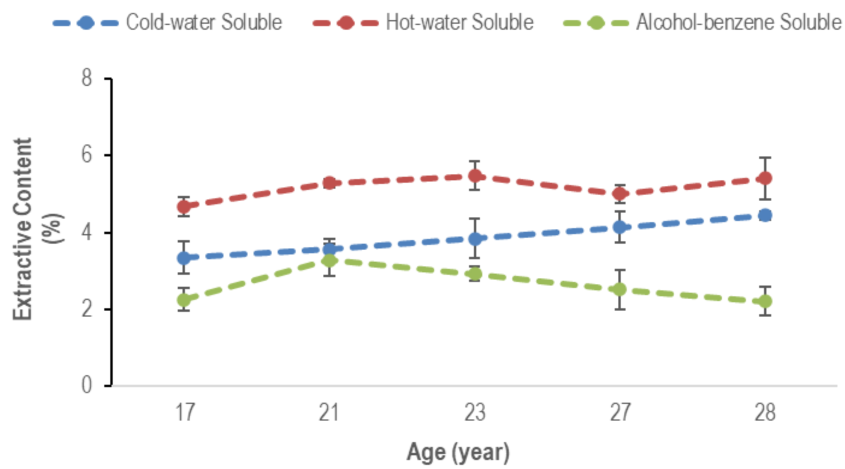


Figure 1. Tusam wood extractive content at various ages

**Tusam Wood Extractive Content**

Wood is a natural material composed of several chemical components, of which one of them is extractive. Wood extractive substances consist of organic and inorganic substances that are included as non-structural components of wood that can be extracted from wood using various solvents such as water, ethanol, acetone, hexane, toluene, and other kinds of solvents (Pattiya 2018). Based on the substance nature, wood extractives can be classified into 4 groups: the volatile extractive group, the soluble extractive group in neutral solvents, the cold-water-soluble extractive group, and the hot-water soluble extractive group. Wood cold-water-soluble extractive generally consists of tannins, gums, simple sugars, and salts or minerals. In contrast, the substances present in hot-water soluble extractives are tannins, gums, simple sugars, salts or minerals, and several phenolic components (Meena and Nimkar 2016). Meanwhile, neutral solvent-soluble extractives such as alcohol-benzene are relatively composed of oleoresin, fat, and wax groups (Nimkar *et al*. 2010). Wood extractives often affect the use and processing of wood (Yoshimoto 1989). Therefore, it is essential to know what types of extractives are contained in the wood to determine the appropriate wood processing technique. In this study, the extractive content of tusam wood at different ages was analyzed. The extractive content analysis was done by determining the cold-water-soluble, hot-water soluble, and alcohol-benzene (1:2) soluble extractive content in tusam wood at different ages.

The content of cold-water extractive, hot-water extractive, and alcohol-benzene extractive was in the range of 3.35-4.45%, 4.68-5.48%, and 2.22-3.28%. The concentration of polar extractive tends to increase as a tree ages, whereas the concentration of non-polar extractive decreases (Rochman *et al*. 2008; Lachowicz *et al*. 2019). Polar extractives are typically contained in cold-water and hot-water extractives. In contrast, non-polar extractives are generally included in neutral solvent-soluble extractives such as alcohol-benzene. The result of cold-water-soluble extractive content and benzene alcohol soluble extractive content measurement in this study was similar to Rachman *et al*. (2008) and Lachowicz *et al*. (2019). However, it was found that hot-water soluble extractive content at all wood ages found in this research was relatively the same. The hot-water soluble content in this research is different from the result obtained by Rachman *et al.* (2008) and Lachowicz *et al.* (2019). The difference can occur because the extractive content in wood is affected by many other factors such as species, growing location, and even the wood storage method and duration (Silvério *et al*., 2008; Chauhan *et al*. 2020).

Table 3. Analysis of variance of the effect of tree age on extractive content.

| | Factor | Cold-Water Soluble Extractive Content | Hot-Water Soluble Extractive Content | Alcohol-benzene Extractive Content |
|---|---|---|---|---|
| Significance (p-value) | Tree age | $2.83 \times 10^{-2}$ * | $7.47 \times 10^{-2}$ ns | $2.58 \times 10^{-2}$* |

Note: * = significant at 95% test level or $p < 0.05$, ns = not significant at 95% test level or $p > 0.05$

### Shear Strength of Tusam Wood Laminate

Compressive shear strength can represent the bonding strength of an adhesive or the bonding quality presented in the composite board. Overall, the adhesive bonding strength can drastically affect the resulting laminate product's quality, such as its stability and mechanical strength. Figure 2 and Figure 3 show the value of the compressive shear strength of tusam wood laminated boards produced in this study. On the laminated board with TGFB adhesive, the shear strength values of the dry blocks respectively ranged from 10.06 – 74.17 kg/cm$^2$, 10.41 – 82.04 kg/cm$^2$, 9.44 – 87.61 kg/cm$^2$, 8.46 – 109.30 kg/cm$^2$ and 8.78 – 79.64 kg/cm$^2$ at wood age of 17 years, 21 years, 23 years, 27 years and 28 years. Meanwhile, for TGFK adhesives, laminated wood boards made from 17 years, 21 years, 23 years, 27 years, and 28 years old wood have a shear strength value of 12.14 – 38.29 kg/cm$^2$, 13.94 – 49.29 kg/cm$^2$. 18.57 – 34.27 kg/cm$^2$, 13.94 – 65.85 kg/cm$^2$ and 12.83 – 37.83 kg/cm$^2$.

All laminated wood in this study met the criteria for compressive shear strength values in dry conditions required by SNI (1998), i.e., 10 kg/cm². In this study, some tusam laminated woods also met the standard of compressive shear strength required by JAS (1998). According to the JAS (1998) standard, the compressive shear strength of laminated wood is ideally around 54 – 96 kg/cm². Laminate wood that successfully met the JAS standard was all laminated wood with TGFB adhesive at 0% extender content, 17 years old laminated wood with TGFB adhesive and 5% extender, and laminated wood made of 27 years old wood with TGFK adhesive and 5% extender.

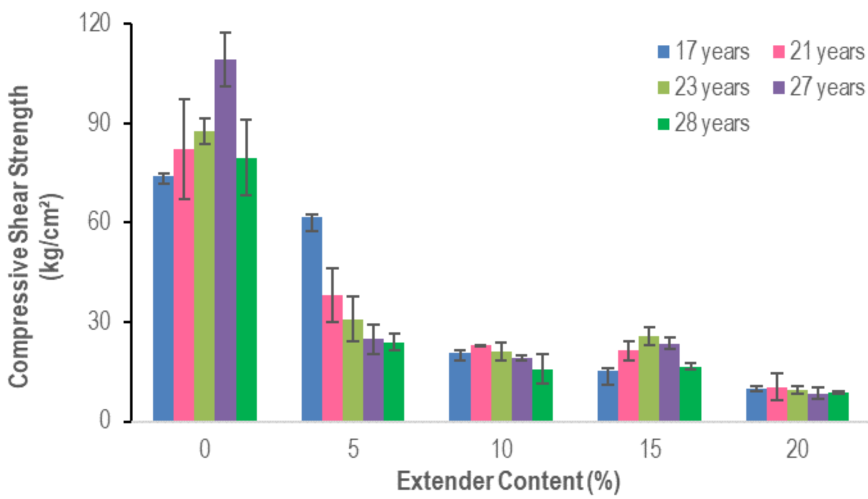


Figure 2. The compressive shear strength of tusam laminated wood in various wood ages bonded using TFGB adhesive with different extender content

The value of gambir tannin adhesive strength obtained in this study was 6-418% higher than gambir tannin adhesive produced by Malrianti *et al*. (2018). In Malrianti *et al*. (2018), the compressive shear strength values of Gambir tannin adhesive laminated board were 11.5 – 21.1 kg/cm². The difference was probably caused by the adhesive manufacturing method used. The compressive shear strength data show that the Gambir tannin adhesive synthesis method employed in this research produced superior results compared to the previous study. The good adhesion strength shown by gambir-TF glue adhesive could be linked to the polymerization reaction between tannin and formaldehyde via the methylene bridge, as stated in prior TF adhesive study. The formation of methylene bridge on TF adhesive can be proved by the formation of a new peak around 2800-2900 cm$^{-1}$ at FTIR spectra of TF adhesive (Rachmawati *et al*. 2018). The average compressive shear strength value in this study was also comparable to the compressive shear strength value on laminated pinewood board bonded with PF adhesive in Júnior (2010) study, which had a value of 37 kg/cm². Previous concern about the lower adhesive power of gambir tannins from PF adhesives due to their solid resin content, which is about 40% lower than PF, did not occur. Some laminated pine wood in this study even obtained a shear strength value of 3-195% greater than PF adhesive laminated wood. In this study, it can be concluded that the solid resin content does not necessarily represent the adhesive strength of an adhesive. A similar result also happens in the research done by Salleh *et al*. (2015). It was found that although the solid content of oil palm starch adhesive (27%) was lower than urea-formaldehyde (UF) adhesive (51%), both adhesives were able to produce rubberwood particleboard with relatively same bonding strength, i.e., ±0.4 MPa at the same adhesive content (15%) and press temperature (165°C) (Salleh *et al*. 2015).

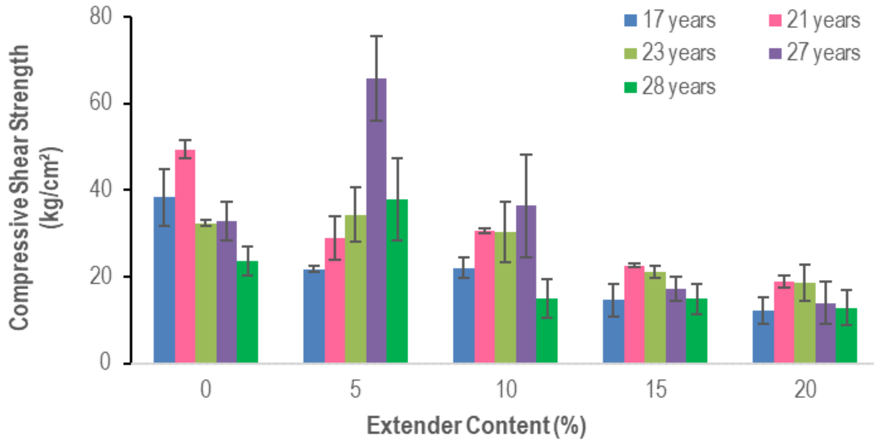


Figure 3. The compressive shear strength of tusam laminated wood in various wood ages bonded using TFGK adhesive with different extender content

Table 4. Analysis of variance in compressive shear strength of laminated wood

| | Factor | Compressive Shear Strength |
|---|---|---|
| Significance (p-value) | Leaf condition | $1.43 \times 10^{-7\,*}$ |
| | Wood age | $1.47 \times 10^{-4\,*}$ |
| | Extender content | $1.83 \times 10^{-42\,*}$ |
| | Leaf condition x Wood age | $5.83 \times 10^{-3\,*}$ |
| | Leaf condition x Extender content | $7.60 \times 10^{-26\,*}$ |
| | Wood age x Extender content | $0.11^{ns}$ |
| | Leaf condition x wood age x Extender content | $1.55 \times 10^{-8\,*}$ |

Note: * = significant at 95% test level or p < 0.05, ns = not significant at 95% test level or p > 0.05

Compared with the adhesive strength of Merbau tannin adhesive in Santoso *et al*. (2016) research, the bonding strength of the gambir tannin adhesive in this study was able to produce 38-145% higher bonding strength. Santoso *et al*. (2016) reported that laminated bamboo with tannin adhesive from Merbau bark extract with 10% resorcinol produced the best compressive shear strength, which was around 44.7 kg/cm$^2$. The adhesive strength of an adhesive itself can be described by the value of the shear strength of the block or the compressive shear strength. According to Réh *et al*. (2021), along with increasing levels of tannins in the adhesive, the strength of the adhesive increases too. The result of gambir tannin adhesive strength in this study which was higher than that of the Merbau bark tannin adhesive in the study of Santoso *et al*. (2016), is an interesting finding. Although the tannin content in gambir leaves in this study was 11-13% lower than Merbau bark tannin content (29%) in the study of Malik *et al*. (2016), they were able to produce better adhesive strength.

The analysis of compressive shear strength results in Table 4 shows that all research factors and interactions between factors except the interaction between wood age and extender content significantly affect the compressive shear strength of laminated wood. From the result of the compressive shear strength test, it is known that gambir leaves with the fresh condition can produce formaldehyde tannin adhesive, which has better bonding strength compared to adhesive from dry leaves. It is presumably due to higher tannin content in fresh leaf extract; hence it can produce an adhesive with better bonding strength.

Meanwhile, the HSD analysis on the age factor showed that the compressive shear strength of the laminated wood at the age of 17, 21, 23, and 27 years was relatively the same. The value of the compressive shear strength of wood at 21, 23, and 27 years was significantly different from that of 28 years old wood. There is a trend of increasing compressive shear strength until a certain age point (from 17 years old to 27 years old), but after that, there is a significant decrease in compressive shear strength (28 years old wood). This might be due to the differences in the basic properties of wood between different ages of wood, for example, the extractive substances. Nazri *et al*. (2009) stated that wood extractive constituents could vary even though the species of wood are the same. One of the factors that influenced it was the difference in the tree's age. If this founding was associated with the results of the extractive content of the wood in this study, it is suspected the extractive compounds found in the cold-water-soluble extractive and benzene alcohol soluble extractive were more significantly affected the bonding quality of tusam wood laminated board.

One interesting thing presented in this research is the effect of alcohol-benzene extractive on laminated tusam board compressive shear strength. In this study, the highest compressive shear strength was produced by wood with the smallest alcohol-benzene soluble extractive content. Generally, the substances contained in the soluble extractive of benzene alcohol or benzene ethanol have properties that interfere with adhesion to wood (Sakuno and Moredo 1998). It is predicted that this case may occur because the extractive effect on the bonding strength may differ depending on the type of adhesive used. For example, in the study of Santoso *et al*. (2019) hot-water extraction treatment on raw materials was able to increase the internal bonding strength of particleboard bonded with citric acid. However, particleboard bonded with sucrose extraction treatment had a negative correlation with the bonding strength. It was suspected that in gambir tannin-based adhesive, the presence of alcohol-benzene extractive substances in tusam wood might have a positive effect on the bonding quality of its laminated composite board.

Based on the findings of the compressive shear strength test, it can also be inferred that the addition of an extender to gambir tannin glue remarkably reduced the bonding strength in this study. The findings of the HSD analysis demonstrate that the compressive shear strength of the adhesive without extender addition is significantly higher than the compressive shear strength of the glue with extender addition. The addition of an extender is intended to reduce adhesive cost and also increase its viscosity and bonding strength. However, in this study, the addition of an extender was not effective in increasing the bonding strength of TF adhesive. This is presumably due to the low tannin content of gambir leaves found in this study. The addition of flour extender in TF adhesive further reduced the amount of tannin substance contained in the adhesive. Tannins relatively give better adhesion strength than starch, which is present in flour. The addition of flour extender in tannin-based adhesive composition decreased the number of compounds with good adhesion ability in the adhesive. This led to the reduced adhesive bonding strength of adhesive; hence it also resulted in the lower compressive shear strength with the addition of an extender.

## Conclusions

The gambir leaves condition that will be extracted, wood age, and addition of extender is known to significantly influence the compressive shear strength of the wood laminate product. The condition of fresh gambir leaves could produce a distinctly higher compressive shear strength value. For the wood age factor, the value of the compressive shear strength tended to increase up to a certain age point (17-27 years), then the value of the shear strength decreased at the age of 28 years. Meanwhile, the addition of an extender in this study had a significant adverse effect on the compressive shear strength.

In general, gambir tannin has good potential to be applied as the wood adhesive for producing laminated tusam wood. This was proved with several gambir tannin formaldehyde adhesive characteristics in this study that could meet the PF standard. In addition, the value of bond strength of laminated tusam wood with gambir tannin formaldehyde adhesive in this study was also able to meet SNI and JAS standards. The value of bonding strength represented by the compressive shear strength produced by laminated products from tusam wood is around 8.46 – 109.30 $kg/cm^2$. The optimum level of extender added to Gambir TF adhesive found in this research was 0%, while the most suitable tusam wood age to produce Gambir TF laminated wood was considered to be 27 years old. Based on its specific gravity, gelation time, and solid content evaluation results, the reactivity of gambir-TF adhesive in this research has not reached its optimum level yet. Additional research is required to determine the treatment that can be applied during the production of gambir-TF adhesive to increase the tannin reactivity and overall features of gambir-TF adhesive. The treatment that can be done includes using different extraction methods to obtain better tannin reactivity or adding a particular compound to facilitate a better polymerization reaction.

## Acknowledgments

We express our deepest gratitude to the Forest Products Research and Development Center for providing the funds and research facilities used to complete this study.

## Author's Contribution

The three authors of this article (AS, ENA, and DAP) were the main contributors to the ideas and experimental designs carried out in this research. AS and ENA did all data collection and data analysis activities. AS, ENA, and DAP carried out the manuscript writing, revision, and finalization.

## References

Abdul Razak, N.; S. Abdul Razak; N. Felix Anak; S. Abdullah. 2014. Effect of initial leaf moisture content on the herbal quality parameter of Orthosiphon stamineus dried leaf during storage. International Journal of Agriculture Innovations and Research 2(6): 1131-1136. ISSN (Online): 2319-1473.

AOAC. Official Methods of Analysis. 18th edition. Association of Official Analytical Chemists; Arlington, VA, USA: 2005.

Atanassova, M.; V. Christova. 2009. Determination of tannins content by titrimetric method for comparison of different plant species. Journal of the University of Chemical Technology and Metallurgy 44 (4): 413-415.

Aulitata, S.; E. Sribudiani; S. Somadona. Karakteristik perekat dan perekatan tanin resorsinol formaldehida pada sirekat akasia (*Acacia mangium*) dan pulai (Alstonia scholaris). Perrenial 17(2): 35-44. p-ISSN: 1412-7784.

Badan Pusat Statistik (BPS). 2021. Luas Area Tanaman Perkebunan Rakyat (Hektar) 2020. Accessed 28 April 2022, https://sumbar.bps.go.id/indicator/54/49/1/luas-area-tanaman-perkebunan-rakyat-.html

Balani, K.; V. Verma; A. Agarwal; R. Narayan. 2014. Physical, Thermal, and Mechanical Properties of Polymer, in : Biosurfaces: A Materials Science and Engineering Perspective, First Edition (Ed. Balani, K., Verma, V., Agarwal, A. and Narayan, R.). John Wiley & Sons, Inc., pp. 329-344. ISBN 9781118950623. DOI: 10.1002/9781118950623.app1.

Basri, E.; T.A. Prayitno; G. Pari. 2012. Pengaruh umur pohon terhadap sifat dasar dan kualitas pengeringan kayu waru gunung (*Hibiscus macrophyllus* Roxb.). Jurnal Penelitian Hasil Hutan 30(4), 243-253. DOI: 10.20886/jphh.2012.30.4.243-253

Blomquist, R.F. 1949. Effect of alkalinity of phenol- and resorcinol resin glues on durability of joints in plywood. Forest Products Laboratory Report No. R1748 (Reaffirmed 1962), Forest Service, U. S. Dept. Agriculture, Madison, Wisconsin, USA.

Chauhan, K.; K.R. Sharma; B. Dutt. 2020. Variation in hot and cold water soluble extractive content in gymnosperms from Western Himalayas. Journal of Pharmacognosy and Phytochemistry 9(5): 1151-1154. DOI: 10.22271/phyto.2020.v9.i5p.12383.

Chen, X.; N. Xia; K. Guo; C. Qi. 2015. Dry bond strength and water resistance of konjac glucomannan, chitosan, and polyvinyl alcohol blend adhesive. Bioresources 10(4): 7038-7052. DOI: 10.15376/biores.10.4.7038-7052.

Departemen Kesehatan Republik Indonesia (Depkes RI). 1995. Farmakope Indonesia. Edisi IV, Cetakan I. Jakarta. 1135-1163.

Dhawale, P.V.; S.K. Vineeth; R.V. Gadhave; J.F.M. Jaffarali; M.V. Supekar; K.M. Thakur; P. Raghavan. 2022. Tannin as a renewable raw material for adhesive applications: a review. Materials Advance 3: 3365-3388. DOI: 10.1039/D1MA00841B

Dongre, P.; M. Driscoll; T. Amidon; B. Bujanovic. 2015. Lignin-furfural based adhesives. Energies 8: 7897-7914. DOI: 10.3390/en8087897

Dunky, M.; A. Pizzi. 2002. Wood Adhesives, in : Adhesion Science and Engineering (Ed. Dillard, D. A., Pocius, A. V. and Chaudhury, M.). Amsterdam, Elsevier Science B.V., pp. 1039-1103. ISBN 9780444511409. DOI: 10.1016/B978-044451140-9/50023-8

Gadhave, R.V.; P.A. Mahanwar; P.T. Gadekar. 2017. Starch-based adhesives for wood/wood composite bonding: review. Open Journal of Polymer Chemistry 7(2): 19-32. DOI: 10.4236/ojpchem.2017.72002.

Garro Gavlez, J.M.; B. Reidl; M. Fechtal. 1996. Gallic acid as a model of tannins in condensation with formaldehyde. Thermochemica Acta 274: 149-163.

Gonultas, O. 2018. Properties of pine bark tannin-based adhesive produced with various hardeners. BioResources 13(4): 9066-9078. DOI:10.15376/biores. 13.4.9066-9078

Griyanitasari, G.; D. Rahmawati; S. Sugihartono; Y. Erwanto. 2019. Cleaner leather tanning process using gambir: the influence of rebating on the properties of leather. Leather and Footwear Journal 19(4): 217-226. DOI: 10.24264/lfj.19.4.6.

Hafiz, N.L.M.; P. Md Tahir; S.H. Lee; Z. Zainal Abidin; F.A. Sabaruddin; N.Y. Mohd Yunus; U.H. Abdullah; H.P.S. Abdul Khalil. 2020. Curing and thermal properties of co-polymerized tannin phenol-formaldehyde resin for bonding wood veneers. Journal of Materials Research and Technology 9(4): 6994–7001. DOI: 10.1016/j.jmrt. 2020.05.029

Hendrik, J.; Y.S. Hadi; M.Y. Massijaya; A. Santoso; A. Pizzi. 2019. Properties of glued laminated timber made from fast-growing species with mangium tannin and phenol resorcinol formaldehyde adhesives. Journal of the Korean Wood Science and Technology 47(3): 253-264. DOI: 10.5658/WOOD.2019.47.3.253

Isnawati, A.; M. Raini; O. Sampurno; D. Mutiatikum; L. Widowati; R. Gitawati. 2012. Karakteristik tiga jenis gambir (*Uncaria gambir* Roxb.) dari Sumatera Barat. Buletin Penelitian Kesehatan 40(4): 201–208.

Japan Plywood Inspection Corporation (JPIC). 2003. Japanese agricultural standard for glued laminated timber. JAS, MAFF. (Notification No. 234: 2003). Tokyo: Japan Plywood

Japanese Agricultural Standard (JAS). 1996. Japanese Agricultural Standar Structural Glued Laminated Timber Notification. No. III. January, 29, 1996. JPIC. Tokyo.

Jeong, S.M.; S.Y. Kim; D.R. Kim; S.C. Jo; K.C. Nam; D.U. Ahn; S.C. Lee. . 2004. Effect of heat treatment on the antioxidant activity of extracts from citrus peels. J. Agric. Food Chem 52: 3389–3393.

Júnior, G.B. 2010. Effects of ply grading and assembly on the properties of plywood panels from Pinus merkusii. Cerne 16(2): 145-153. DOI: 10.1590/S0104-77602010000200005

Kanto, K.; A.P. Kusumadewi; S. Sutjipto. 2008. Pengaruh waktu pengeringan terhadap kadar tanin daun Jati Belanda (*Guazuma ulmifolia* Lamk.). Jurnal Tumbuhan Obat Indonesia 1(1): 38-46.

Karliati, T.; F. Febrianto; W. Syafii, I. Wahyudi; I.N.J. Wistara. 2014. Gutta-percha-based adhesive for laminated wood production. BioResources 9(3): 5034-5044. DOI: 10.15376/biores.9.3.5034-5044

Kassim, M.J.; M.H. Hussin; A. Achmad; N.H. Dahon; T.K. Suan; H.S. Hamdan. 2011. Determination of total phenol, condensed tannin and flavonoid contents and antioxidant activity of Uncaria gambir extracts. Indonesian Journal of Pharmacy: 50-59. DOI: 10.14499/INDONESIANJPHARM0ISS0PP50-59

Kementerian Pertanian (Kementan). 2020. Rencana Strategis Kementerian Pertanian 2020-2024. Jakarta, Kementrian Pertanian, pp. 134.

Khiari, R.; N. Baaka; M. Ammar; M.K. Saad. 2017. Properties of tannin-glyoxal resins prepared from lyophilized and condensed tannin. Journal of Textile Engineering & Fashion Technology 3(4): 00110. DOI: 10.15406/jteft.2017.03.00110.

Kim, H.Y. 2015. Statistical notes for clinical researchers: Type I and type II errors in statistical decision. Restorative Dentistry & Endodontics 40(3): 249–252. DOI: 10.5395/rde.2015.40.3.249.

Kurnia, V.C.; S. Sumiyati; G. Samudro. 2017. Pengaruh kadar air terhadap proses pengomposan sampah organik dengan metode open windrow. Jurnal Teknik Mesin 6(2): 119-123. DOI:10.22441/jtm.v6i2.1191

Lachowicz, H.; H. Wróblewska; R. Wojtan; M. Sadjak. 2019. The effect of tree age on the chemical composition of the wood of silver birch (*Betula pendula* Roth.) in Poland. Wood Science and Technology 53: 1135–1155. DOI: 10.1007/s00226-019-01121-z.

Lee, S.; D.K. Lee. 2018. What is the proper way to apply the multiple comparison test? Korean Journal of Anesthesiology 71(5): 353–360. DOI: 10.4097/kja.d.18.00242

Makkar, H.P.S.; B. Singh; S.S. Negi. 1990. Tannin levels and their degree of polymerisation and specific activity in some agro-industrial by-products. Biological Wastes 31(2): 137–144. DOI: 10.1016/0269-7483(90)90167-q

Malik, J.; A. Santoo; Y. Mulyana; B. Ozarska. 2016. Characterization of Merbau Extractives as a Potential Wood-Impregnating Material. BioResources 11(3): 7737–7753. DOI: 10.15376/biores.11.3.7737-7753.

Malrianti, Y.; A. Kasim; N. Novelina. 2018. Tannins and catechins content of gambier (*Uncaria gambier* Roxb) in relation with adhesive qualities and bonding strength of cold setting glue. International Journal of Advanced Research 6(12): 622-627. DOI: 10.21474/IJAR01/8181

Mamza, P.A.P.; E.C. Ezeh; E.C. Gimba; D.E. Arthur. 2014. Comparative study of phenol formaldehyde and urea formaldehyde particleboards from wood waste for sustainable environment. International Journal of Scientific & Technology Research 3(9): 53-61. ISSN 2277-8616.

Martín-Cabrejas, M.A.; Y. Aguilera; M. Pedrosa; C. Cuadrado; T. Hernández; S. Díaz; R. Esteban. 2009. The impact of dehydration process on antinutrients and protein digestibility of some legume flours. Food Chem. 114: 1063-1068.

McHugh, M.L. 2011. Multiple comparison analysis testing in ANOVA. Biochemia Medica 21(3): 203-209. DOI: 10.11613/BM.2011.029

Meena, R.K.; A.U. Nimkar. 2016. Variation in soluble extractives, lignin and holocellulose content of wood of different provenances of Tectona grandis L. Journal of Applied and Natural Science 8(1): 80-83. DOI: 10.31018/JANS.V8I1.751

Miranda, M.; A. Vega-Gálvez; J. López; G. Parada; M. Sanders; M. Aranda; E. Uribe; K. Di Scala. 2010. Impact of air-drying temperature on nutritional properties, total phenolic content and antioxidant capacity of quinoa seeds (*Chenopodium quinoa* Willd.). Industrial Crops and Products 32(3): 258-263. DOI: 10.1016/j.indcrop.2010.04.019

Müller, C.; U. Kües; K. Schöpper; A. Kharazipour. 2007. Natural binders, Wood Production, Wood Technology, and Biotechnological Impacts (Ed. U. Kües). Göttingen, Universitätsverlag Göttingen, pp.347-381.

Nazri, W.M.; K. Jamaludin; M.N. Rudaini; S. Rahim; M.Y. Nor Yuziah. 2009. Effects of chemical components on properties of oriented strand board from *Leucaena leucocephala* wood. Journal of Tropical Forest Science 21(4): 353-360.

Nimkar, A.U.; K.R. Sharma; S.A. Nimkar. 2010. Variation in soluble extractives, lignin and holocellulose content of wood high resin yielders and check trees in Chirpine (*Pinus roxburghii* Sargeant). Journal of Nontimber Forest Products 17 (2): 227-231.

Pattiya, A. 2018. Fast pyrolysis, in: Direct Thermochemical Liquefaction for Energy Applications. (Ed. Rosendahl, L.). Swaston, Woodhead Publishing, pp. 3-24. ISBN: 978-0-08-101029-7.

Pizzi; A.; H.O. Scharfetter. 1978. The chemistry and development of tannin-based adhesives for exterior plywood. Journal of Applied Polymer Science 22(6): 1745-1761. DOI: 10.1002/app.1978.070220623.

Pizzi, A. 2006. Recent developments in eco-efficient bio-based adhesives for wood bonding: Opportunities and issues. Journal of Adhesion Science and Technology 20(8): 829-846. DOI: 10.1163/156856106777638635.

Pizzi, A. 2008. Tannins: Major Sources, Properties and Applications, in: Monomers, Polymers and Composites from Renewable Resources. (Ed. Belgacem, N. M.). Elsevier, pp. 179-199. ISBN 9780080453163. DOI: 10.1016/B978-0-08-045316-3.00008-9.

Pizzi, A. 2016. Wood products and green chemistry. Ann. For. Sci. 73(1): 185-203. DOI: 10.1007/s13595-014-0448-3

Rachman, O.; N. Hadjib; J. Jasni; A. Santoso; G. Pari; S. Rulliaty; J. Malik. 2008. Penetapan Daur Teknis Kayu HTI Sengon untuk Bahan Baku Kayu Pertukangan. Laporan Tahunan 2008. Bogor: Pusat Penelitian dan Pengembangan Hasil Hutan.

Rachmawati, O.; P. Sugita; A. Santoso. 2018. Sintesis perekat tanin resorsinol formaldehida dari ekstrak kulit

pohon Mangium untuk peningkatan kualitas batang sawit. Jurnal Penelitian Hasil Hutan 36(1): 33-46. DOI: 10.20886/jphh.2018.36.1.33-46
Raydan, N.D.V.; L. Leroyer; B. Charier; E. Rbles. 2021. Recent advances on the development of protein-based. Molecules 26: 7617. DOI: 10.3390/molecules 26247617.
Réh, R.; L. Krišťák; J. Sedliačik; P. Bekhta; M. Božiková; D. Kunecová; V. Vozárová; E.M. Tudor; P. Antov; V. Savov. 2021. Utilization of birch bark as an eco-friendly filler in urea-formaldehyde adhesives for plywood manufacturing. Polymers 13(4): 511. DOI: 10.3390/ polym13040511
Sakuno, T.; C. Moredo. 1998. Bonding Properties of Some Tropical Woods after Solvent Extraction. Proceeding of the second International Wood Science Seminar, Serpong, Indonesia. pp. 183-189.
Salleh, M. K.; R. Hashim; O. Sulaiman; S. Hiziroglu; W.N.A. Wan Nadhari; N. Abd Karim; N. Jumhuri; L. Ang. 2015. Evaluation of properties of starch-based adhesives and particleboard manufactured from them. Journal of Adhesion Science and Technology 29(4): 319–336. DOI: 10.1080/01694243.2014.987362.
Saminathan, M.; H. Tan; C. Sieo; N. Abdullah; C. Wong; E. Abdulmalek; Y. Ho. 2014. Polymerization degrees, molecular weights and protein-binding affinities of condensed tannin fractions from a *Leucaena leucocephala* hybrid. Molecules 19(6): 7990–8010. DOI: 10.3390/molecules19067990.
Santoso, A. 2003. Komposisi Resin dan Kadar Adiktif dalam Perekat Lignin Resorsinol Formaldehida pada Kayu Lamina Kempas, Jurnal Teknologi Hasil Hutan, Institut Pertanian Bogor 16(2), Bogor.
Santoso, A; Y.S. Hadi; J. Malik. 2012. Tannin resorcinol formaldehyde as potential glue for the manufacture of plybamboo. Indonesian Journal of Forestry Research 9(1): 10-15. DOI: 10.20886/ijfr.2012.9.1.10-15
Santoso, A.; I.M. Sulatiningsih; G. Pari; J. Jasni. 2016. Pemanfaatan untuk ekstrak kayu merbau perekat produk laminasi bambu. Jurnal Penelitian Hasil Hutan 34(2): 89-100.
Santoso, A.; A. Abdurachman. 2016. Karakteristik ekstrak kulit kayu mahoni sebagai bahan perekat kayu. Jurnal Penelitian Hasil Hutan 34(4): 269-284. DOI: 10.20886/jphh.2016.34.4.269-284
Santoso, M.; R. Widyorini; T.A. Prayitno; J. Sulistyo. 2019. The effects of extractives substances for bonding performance of three natural binder on nipa fronds particleboard. KnE Life Sciences 4(11): 227-238. DOI:10.18502/kls.v4i11.3868
Silvério, F.O.; L.C.A. Barbosa; C.R.A. Maltha; P.H. Fidêncio; M.P. Cruz; D.P. Veloso; A.F. Milanez. 2008. Effect of storage time on the composition and content of wood extractives in Eucalyptus cultivated in Brazil. Bioresource Technology 99(11): 4878-4886. DOI: 10.1016/j.biortech.2007.09.066
Standar Nasional Indonesia (SNI). 1989a. Cara Uji Kadar Sari Ekstrak (Alkohol- Benzena) dalam Kayu. Badan Standarisasi Nasional. Jakarta.
Standar Nasional Indonesia (SNI). 1989b. Cara Uji Kelarutan Kayu dan Pulp dalam Air Dingin dan Air Panas. Badan Standarisasi Nasional. Jakarta.
Standar Nasional Indonesia 1998, Fenol formaldehida cair untuk perekat kayu lapis (SNI 06-4567-1998), Standar Nasional Indonesia, Jakarta.
Standar Nasional Indonesia 1999, Kualitas perekatan untuk perekatan kayu lapis (SNI 06-6049-1999), Standar Nasional Indonesia, Jakarta.
Stauffer, E.; J.A. Dolan; N. Reta. 2008. Chemistry and Physics of Fire and Liquid Fuels, in : Fire Debris Analysis. (Ed. Stauffer, E., Dolan, J.A. and N., Reta). Academic Press, pp. 85-129. ISBN 9780126639711. DOI: 10.1016/B978-012663971-1.50008-7.
Stewart, J.L.; F. Mould; I. Mueller-Harvey. 2000. The effect of drying treatment on the fodder quality and tannin content of two provenances of *Calliandra calothyrsus* Meissner. Journal of the Science of Food and Agriculture 80(10): 1461-1468. DOI:10.1002/1097-0010(200008)80:10<1461::AID-JSFA672>3.0.CO;2-R.
Sucipto, T.; S. Ruhendi. 2012. Analisis kualitas perekatan kayu laminasi mangium dengan perekat polistirena. FORESTA Indonesian Journal of Forestry 1(1), 19-24. ISSN: 2089-9890
Sucipto, T.; R. Widyorini; T.A. Prayitno; G. Lukmandaru. 2020. Properties of a new adhesive composed of gambir-sucrose. Journal of the Korean Wood Science and Technology 48(3): 303-314. DOI: 10.5658/WOOD. 2020.48.3.303
Umemura, K.; S. Hayashi; S. Tanaka; K. Kanayama. 2017. Changes in physical and chemical properties of sucrose by the addition of ammonium dihydrogen phosphate. Journal of The Adhesion Society of Japan 53(4): 112-117. DOI:1 0.11618/adhesion.53.112
Wangaard, F.F. 1946. Summary of information on the durability of woodworking glues. Forest Products Laboratory Report No. 1530 (Revised 1956 as: Durability of water-resistant woodworking glues), Forest Service, U. S. Dept. Agriculture, Madison, Wisconsin, USA.
Xing, C.; S.Y. Zhang; J. Deng; S. Wang. 2006. Urea–formaldehyde-resin gel time as affected by the ph value, solid content, and catalyst. Journal of Applied Polymer Science 103(3): 1566-1569. DOI:10.1002/app. 25343
Yoshimoto, T. 1989. Effect of extractives on the utilization of wood, in: Natural products of woody plants II – Chemical extraneous to lignocellulosics cell wall. (Ed. Rowe, J. W). Berlin, Springer-Verlag, pp. 920-931.
Zanetti, M.; V. Causin; R. Saini; A. Cardin; R. Cavalli. 2014. Effect of tannin on increasing UF adhesive performance at high temperature investigated by TMA and TGA analysis. European Journal of Wood and

Wood Products 72(3): 385–392. DOI: 10.1007/s00107-014-0795-7
Zhang, L.H.; S.J. Zhang; G.F. Ye; H.B. Shao; G.H. Lin; M. Brestic. 2013. Changes of tannin and nutrients during decomposition of branchlets of Casuarina equisetifolia plantation in subtropical coastal areas of China. Plant Soil Environment 59(2): 74-79. DOI:10.17221/598/2012 -PSE.
Zhang, T.; C. Yu; M. Yu; Y. Huang; J. Tan; M. Zhang; X. Zhu. 2022. Multifunctional tannin extract-based epoxy derived from waste bark as a highly toughening and strengthening agent for epoxy resin. Industrial Crops and Products 176: 114255. DOI: 10.1016/j.indcrop. 2021.114255
Zhou, X.; G. Du. 2019. Applications of tannin resin adhesives in the wood industry, Tannins - Structural Properties, Biological Properties and Current Knowledge in: (Ed. A. Aires). London, IntechOpen. DOI: 10.5772/intechopen.86424.

Adi Santoso
Research Center of Biomass and Bioproducts,
Research Organization for Life Sciences and Environment,
National Research and Innovation Agency (BRIN),
Jl. Raya Jakarta-Bogor km. 46, Cibinong,
Bogor 16911, Jawa Barat
E-mail : profadisantoso@gmail.com

Erlina Nurul Aini
Research Center of Biomass and Bioproducts,
Research Organization for Life Sciences and Environment,
National Research and Innovation Agency (BRIN),
Jl. Raya Jakarta-Bogor km. 46, Cibinong,
Bogor 16911, Jawa Barat
E-mail : ainierlinanurul@gmail.com
erlina.nurul.aini@brin.go.id

Dina Alva Prastiwi
Cilegon College of Analytical Chemistry
Jl. KH. Wasyid No. 6 Jombang Wetan
Cilegon 42411, Banten
E-mail : dinaalvastakc@gmail.com