

# Utilization of Tannin Based Adhesive from Mahogany (Swietenia Mahagony) Extract Bark for Composite Product

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## Research Article

**Keywords:** Swietenia mahagoni, tannin extract, adhesive, plywood, Acacia mangium, Paraserianthes falcataria

**Posted Date:** November 23rd, 2022

**DOI:** <https://doi.org/10.21203/rs.3.rs-2292486/v1>

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# Abstract

The aim of this study was to determine the quality of mahogany (*Swietenia mahagony*) tannin extract as an adhesive for plywood. The mahogany tannin extract was copolymerized with phenol and formaldehyde to become TPF adhesive. Then, the plywood was made from mangium (*Acacia mangium*) and sengon (*Paraseriathes Falcataria*) with dimensions  $30 \times 30 \times 0.6$  (cm<sup>3</sup>) in the length, width, and thickness, respectively. Pressing was carried out using the cold press method for 11 minutes and then followed by the hot press method for 10 minutes at a temperature of 110 °C with a specific pressure of 1.47 MPa. Phenol formaldehyde and urea formaldehyde adhesives were used as a comparison and 5 replicates were made for each plywood. The physical and mechanical properties were tested according to JAS 233:2003. The result showed that TPF had an equal quality with the synthetic adhesive in dry conditions. Moreover, all of plywood from sengon wood categorized as F\*\*\*\* the lowest and the best.

## 1. Introduction

Diversification of the use of wood originating from plantation forests is one of the methods to meet the community's timber needs. This is caused by the increase in demand for wood which is no longer in line with the availability of wood in natural forests. Timber originating from plantation forests has a relatively short harvest cycle, ranging from 5–10 years (Hadi et al. 2020). In short, the harvest cycle causes wood from plantation forests to have a higher sapwood content than heartwood. In addition, plantation forest wood has a diameter of less than 30 cm, high defects, and low durability, making it easily attacked by destructive organisms. In order to meet the community's need for large-diameter wood, this fast-growing wood species is used as an alternative to natural forest wood through the manufacture of engineered wood products such as laminated products (Lestari et al. 2018).

Laminate product industries such as plywood generally still use synthetic adhesives such as polyurethane, methylene diphenyl diisocyanate (MDI), phenol-resorcinol-formaldehyde (PRF), and urea formaldehyde (UF) (Sun et al. 2019). The synthetic adhesive raw materials come from non-renewable resources such as fossils and petroleum. As a result, over time the availability of synthetic raw materials in nature will decrease. The diminishing sources of oil and unstable oil prices will lead to uncertain future costs and availability of synthetic adhesives (Kusumah et al. 2017). High-priced synthetic adhesives will increase the final cost of the wood panels produced (Prabuningrum et al. 2020). In addition, the use of synthetic adhesives has a high risk to humans and the environment (Widyorini et al. 2019). Along with increasing problems in the gluing industry in recent years, the manufacture of adhesives from natural materials that are renewable and environmentally friendly has begun to become the target of intensive research by world researchers.

## 2. Material And Methods

### Mahogany Tannin Adhesive Formulations

Tannins that have been formed into powder are added to distilled water with a ratio of 1: 3 (w/w). Mahogany tannin adhesive has two components, which consist of resin (tannin, phenol, and 40% NaOH) and 37% formaldehyde (as a crosslinker). Tannin (T) is mixed with phenol (R) and then NaOH is added until it reaches a pH of 11. Formaldehyde (F) is added to the TP mixture with a ratio of 100:5 based on weight, then stirred until homogeneous.

#### Characterization of mahogany tannin adhesive

The physical properties of TRF adhesive include solids content, viscosity, visual appearance, and gelation time. Characterization of the chemical properties of the mahogany tannin adhesive, namely determining the pH, and determining the chemical components using py-GCMS.

#### Plywood manufacturing

Mahogany tannin adhesive was applied to finir sheets from 2 types of wood, namely mangium (*Acacia mangium*) and sengon (*Paraseriathes Falcataria*) to form a plywood product (with a total of 3 layers) measuring  $30 \times 30 \times 0.6$  (cm<sup>3</sup>) in the dimensions of length, width, and thickness. Pressing was carried out using the cold press method for 11 minutes and then followed by the hot press method for 10 minutes at a temperature of 110 °C with a specific pressure of 1.47 MPa. Phenol formaldehyde and urea formaldehyde adhesives were used as a comparison and 5 replicates were made for each plywood. After going through the pressing process, the plywood was air-dried for  $\pm 14$  days before testing its physical and mechanical properties with relative humidity ranging from 60% – 70% and room temperature (25 °C – 32 °C).

#### Physical and Mechanical Properties of Plywood

Plywood characterization includes physical and mechanical properties according to JAS 233–2003. Physical properties include moisture content and density, mechanical properties include tensile shear strength. In addition, delamination tests and formaldehyde emission tests were also carried out.

## 3. Result And Discussions

#### Physical and Chemical Properties of Mahogany Tannin Adhesive

Mahogany tannin (TPF) adhesive had a liquid phase, dark-brown color, and smell like phenol. Viscosity of TPF adhesive was 2.1 poise and its more liquid than the PF adhesive 30 poise. The solid content of TPF was 27.27 and had a gelatinous time 120 minutes.

The py-GCMS of TPF adhesive results shown in the chromatograph (Figure 1) indicate that the mahogany tannin adhesive contains several phenolic compounds. At a retention time of 16.123 minutes and at a retention time of 16.292 minutes were a phenolic compound o-Cresol with a total concentration of 9.29%, at a retention time of 16.517 minutes is Phenol, 4-methyl- (CAS) p-Cresol with a concentration of 10.79%, at a retention time of 17.163 minutes is a Phenol, 2,4-dimethyl- (CAS) 2,4-Xylenol compound

with a concentration of 5.91%, and Phenol, 2,3,6-trimethyl- (CAS) 2,3,6-Trimethylphenol at a retention time of 17.525 minutes with a concentration of 0.77%. the total phenolic compound of TPF adhesive was 26.76%.

## Performance of Plywood

The physical and mechanical performance of plywood bonded with TPF adhesive were evaluated according to JAS 233:2003.

**Table 2. Physical properties of plywood**

| Type of Adhesive      | Type of Wood | Physical                      |                           | Delamination (%)<br>** |
|-----------------------|--------------|-------------------------------|---------------------------|------------------------|
|                       |              | Density (g·cm <sup>-3</sup> ) | Moisture content (%)<br>* |                        |
| TPF                   | Mangium      | 0.61 (0.03)                   | 12.7 (0.9)                | 100.0 ± 0.0            |
|                       | Sengon       | 0.31 (0.04)                   | 11.1 (0.3)                | 100.0 ± 0.0            |
| PF                    | Mangium      | 0.67 (0.02)                   | 12.5 (0.4)                | 0.0 ± 0.0              |
|                       | Sengon       | 0.32 (0.01)                   | 11.4 (0.3)                | 0.0 ± 0.0              |
| UF                    | Mangium      | 0.64 (0.02)                   | 12.2 (0.3)                | 100.0 ± 0.0            |
|                       | Sengon       | 0.32 (0.01)                   | 11.8 (0.3)                | 100.0 ± 0.0            |
| JAS Standard 233-2003 |              |                               | Max 14                    |                        |

\*Maximum moisture content was 14%, \*\*maximum non-delaminated part was less than 50 mm (delamination < 33.3%).

The physical properties, density and moisture content, of plywood are shown in Table 2. The density of plywood which was made from each of the three adhesives and mangium wood had the same density, as well as that of sengon plywood which was bonded using each of the three types of adhesives. This is also evidenced by the results of the analysis of variance (Table 3), which shows that the density produced using the three types of adhesives is not significantly different between plywood. It can be caused by a similar thickness of veneers used. In general, there is not significantly different density among the plywood (Suhasman and Agussalim, 2019).

The moisture content of all of plywood ranged from 11.1%-12.7%, these values reached equilibrium moisture content 10.6%-20.5% (Simpson 1991), and also fulfilled the JAS 233-2003 standard (below 14%). Based on the analysis of variance (Table 2), neither the wood species nor the material affected the moisture content. Veneer and plywood samples for each wood species were conditioned for a month, and the moisture content of the products approached the equilibrium moisture content in Makassar.

Delamination is an important characteristic of plywood, especially when it is intended for outdoor applications, i.e., exposed to cyclic changes of MC. These changes lead to the formation of mechanical stresses in the glue joint, resulting from the differences in shrinking and swelling of adjacent veneer layers due to the varied MC. The TPF and UF adhesives used in this work exhibited poor wet bonding strength, with a delamination value of 100%. By contrast, plywood using PF showed 0% of delamination, showing that PF resins had superior wet bonding strength. The delamination of plywood bonded with TPF and UF did not meet the minimum requirement of JAS 233-2003 with a minimum of 50 mm non-delaminated portion or lower than 33.3%. This represents a serious drawback for the industrial application of the tested especially TPF adhesives compared to the PF wood adhesives. The chemical modification of veneers is a viable approach to increase the dimensional stability of plywood in order to reduce its delamination and deformation. Similar research results were obtained by Lubis et al. (2022), where the natural lignin adhesive (100%) has a delamination value that contrasts with the PU synthetic adhesive used (0%).

The tensile shear strength value of plywood in Figure 2 shows that all plywood meets the JAS 233-2003 standard. In order of value, the plywood derived from sengon wood was higher than that of mangium wood for the three types of adhesives. Mangium wood has a higher specific gravity than sengon wood, so that sengon has a higher porosity than mangium (Karlinasari et al. 2018). Priadi et al (2019) also stated that the more porous a wood is, the greater the ability of the adhesive to penetrate and form anchors, causing a stronger adhesive bond to form.

Analysis of variance (Table 3) shows that there is no significant difference between the types of TPF, PF, and UF adhesives on the quality of the tensile shear strength of the resulting plywood. While the types of wood, sengon wood and mangium wood have very significant differences.

**Table 3. ANOVA of the physical and mechanical properties of plywood.**

| Parameter              | Wood Species | Adhesive |
|------------------------|--------------|----------|
| Density                | **           | NS       |
| Moisture content       | **           | NS       |
| Tensile shear strength | **           | NS       |
| Delamination           | NS           | **       |

\*Signifikan (p-value<0.05)

\*\*Sangat Signifikan (p-value<0.01)

NS = Not Significant

### Formaldehyde Emission

**Table 4. Emisi Formaldehida plywood.**

| Type of wood | Formaldehyde Emission of Adhesive (%) |      |      | Classification <sup>*)</sup> |
|--------------|---------------------------------------|------|------|------------------------------|
|              | TPF                                   | PF   | UF   |                              |
| Mangium      | 0.45                                  | 0.25 | 0.65 | F***                         |
| Sengon       | 0.05                                  | 0.01 | 0.07 | F****                        |

<sup>\*)</sup> Categorized by JAS 233-2003

Adhesives that use formaldehyde will emit formaldehyde into the air. This emission occurs because the formaldehyde does not react completely which is eventually released into the environment. This happens because the heating point in formaldehyde is below the temperature. Formaldehyde emission at certain levels will cause several health problems such as dizziness, nausea and watery eyes. Table 6 shows that the formaldehyde emission values of mangium plywood using TPF, PF, and UF adhesives are included in the F\*\*\* classification, and sengon plywood with all three types of tannin adhesives is included in the F\*\*\*\* classification as the lowest and best emission categories according to JAS standard 233-2003. This result was similar to Lestari et al. (2019), who tested formaldehyde emissions on glulam product using mahogany tannin extract copolymerize with resorcinol and formaldehyde.

## Conclusion

The result of these research shows mahogany tannin adhesive or TPF contain 15,7% of phenolic compound. Moreover, all of plywood fullfild the JAS 233-2003 standard for moisture content and tensile shear strength. Not only moisture content and tensile shear strength, but also for formaldehyde emission test all of plywood from sengon wood categorized as F\*\*\*\* the lowest and the best. However, TPF and UF did not fullfild the standard in the delaminations test. It could be concluded that TPF had an equal quality with the synthetic adhesive in dry conditions.

## Declarations

## ACKNOWLEDGEMENT

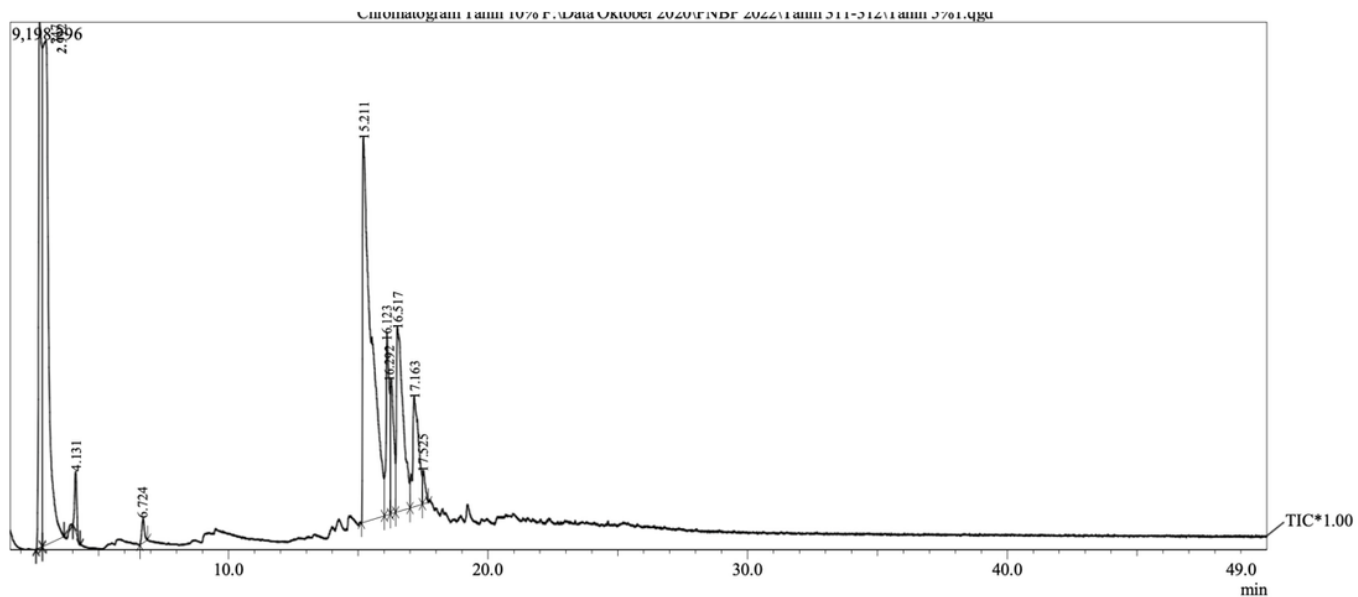
We acknowledge the sponsorship of the Hasanuddin University for conducting this research through excellent research of Hasanuddin University research grant scheme 2022.

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## Figures



| Retention time (min) | Name of Compound                                     | Concentration (%) |
|----------------------|------------------------------------------------------|-------------------|
| 16.123               | Phenol, 2-methyl- (CAS) o-Cresol                     | 5,42              |
| 16.292               | Phenol, 2-methyl- (CAS) o-Cresol                     | 3,87              |
| 16.517               | Phenol, 4-methyl- (CAS) p-Cresol                     | 10.79             |
| 17.163               | Phenol, 2,4-dimethyl- (CAS) 2,4-Xylenol              | 5,91              |
| 17.525               | Phenol, 2,3,6-trimethyl- (CAS) 2,3,6-Trimethylphenol | 0.77              |

**Figure 1**

Py-GCMS chromatograph of mahogany tannin adhesive

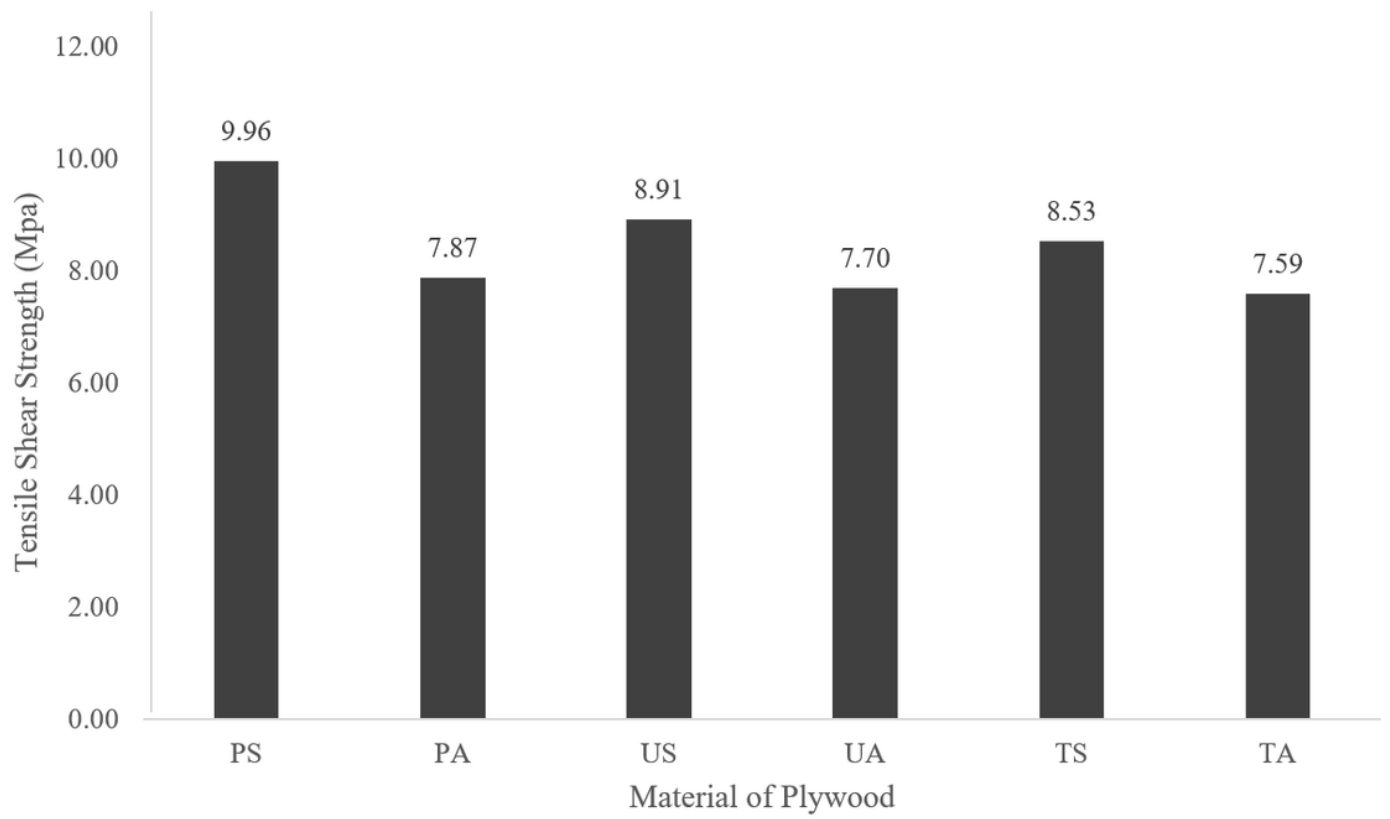


**Figure 2**

**Dry bonding strength of Plywood**