Effect of Lignin or Lignosulfonate Addition on the Fire Resistance of Areca (Areca catechu) Particleboards Bonded with Ultra-Low-Emitting Urea-Formaldehyde Resin

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Abstract: As a way to accommodate the rising demand for “green” wood-based products, agricultural waste from Areca (Areca catechu) nut farms, which is generally burned on-site, can be used to raise the value of alternative lignocellulosic raw materials. This research aimed to investigate and evaluate the effect of technical lignin (kraft lignin or lignosulfonate) addition on particleboard properties from areca bonded with ultra-low-emitting urea formaldehyde (UF) resin. The physical properties, mechanical properties, and fire resistance of the laboratory-made particleboards were tested and evaluated in accordance with the applicable Japanese industrial standards (JIS). The highest density of 0.84 g/cm³ was determined for the laboratory boards, bonded with an adhesive mixture of UF resin and kraft lignin with three washing treatments. The lowest moisture content of 9.06%, thickness swelling of 71.16%, and water absorption of 129.17% were determined for the boards bonded with lignosulfonate with five washing treatments, with commercial lignin, and with lignosulfonate with five washing treatments, respectively. The highest MOR and MOE values, i.e., 113.49 kg/cm² and 10,663 kg/cm², respectively, were obtained for the particleboards bonded with lignosulfonate with five washing treatments. Interestingly, all laboratory boards exhibited good fire resistance following the UL-94 standard. Based on the gas torch test, the lowest weight loss of 16.7% was determined in the boards fabricated with lignosulfonate with five washing treatments. This study demonstrated that adding lignin-based fire retardants represents a viable approach to producing lignocellulosic composites with enhanced fire resistance and a lower carbon footprint.

Keywords: fire retardant; lignin; lignosulfonate; areca nut sheath; particleboard; ultra-low emission UF

1. Introduction

Areca (Areca catechu) nuts are one of the plantation crops that are grown extensively in Indonesia, particularly in Sumatra, Sulawesi, Kalimantan, and Papua [1]. Areca nut leaf sheaths (ALS) have a high amount of cellulose, yet they are frequently discarded as waste material and are easily available for free or at a very low cost [2]. Agricultural waste from areca nut farms is estimated to be approximately 8 million tonnes of leaf sheaths per year [3]. Most of this agricultural biomass is not reused in value-added applications...
but is typically burned on-site for disposal because it is less expensive than other options. Burning agricultural waste has an adverse impact on both local flora and fauna and on human health (respiratory disorders), as well as increasing the possibility of fire spread and greenhouse gas emissions [4].

Wood is the most popular lignocellulosic natural feedstock for manufacturing paper pulp, furniture products, building materials, and fuel. Due to the scarcity of wood resources, the efficient industrial utilization of lignocellulosic agricultural biomass in manufacturing particleboards and other wood-based composites will support the transition of the wood sector towards the adoption of a circular bioeconomy, resulting in minimized waste generation and environmental pollution [4]. Particleboards are wood composite materials manufactured from wood or other lignocellulosic particles bonded with thermosetting polymeric adhesive at specific pressures and temperatures [5].

The most commonly used synthetic adhesives for bonding wood composites are based on formaldehyde, e.g., urea-formaldehyde (UF), melamine formaldehyde (MF), phenol-formaldehyde (PF), and other thermosetting resins, representing nearly 95% of the total adhesives used in the wood industry [6,7]. UF resins are the most widely used wood adhesives, with an estimated consumption of 11 million tons per year worldwide due to their superior adhesion capabilities, high reactivity, solubility in water, short press times, low curing temperatures, comparably inexpensive costs, and simplicity of handling. However, these aminoplastic resins are characterized by severe drawbacks, such as inferior water resistance and hazardous free formaldehyde emission from the composites, associated with serious adverse human health effects, including cancer [8–11]. As a result, this study was planned to synthesize and use an ultra-low-emitting formaldehyde UF resin with a formaldehyde to urea (F/U) ratio of 0.8.

Lignin is the second most abundant natural polymer found in a variety of raw materials, including woody biomass and agricultural waste. In recent decades, views on lignin have shifted from being regarded as a waste material used as animal feed and low-grade fuel to a promising natural feedstock for value-added applications such as polymers, adhesives [12], coatings [13], and fire retardants [14]. The use of lignin-based fire-retardant materials has the advantage of being environmentally friendly because they come from biomass [15,16]. In composites, lignin can be utilized as a filler material, a nucleating agent, a compatibilizer, and a coupling agent by combining it with synthetic and natural polymers. The prospective applications of composite properties may impact various processes and sources [17]. Lignin isolated from black liquor with different washing frequencies and synthesizing lignosulfonate can enhance the material’s thermal and fire-resistant characteristics [14]. Lignosulfonates, i.e., sulfonated lignin, synthesized by using sulphurous acid and either sulfite or bisulfite salts containing magnesium, sodium, calcium, or ammonium at varying pH levels, are the most abundant and commercially available technical lignin worldwide [18,19].

This research work aimed to investigate and evaluate the effect of technical lignin (kraft lignin or lignosulfonate) addition on the fire resistance, physical, and mechanical properties of particleboards manufactured from areca and bonded with ultra-low-emitting formaldehyde (ULEF) UF resin.

2. Materials and Methods

The materials used in this research were commercial lignin (catalogue number: 370959) with a purity content and transition glass of 90.34% and 141 °C, commercial lignosulfonate (catalogue number: 471038) with a purity content and Tg of 16.19% and 158 °C from Sigma-Aldrich, Eucalyptus kraft-lignin from black liquor with 3 and 5 washing treatments, synthesized lignosulfonate from isolated Eucalyptus lignin, NaOH in analytical grade by Merck (Darmstadt, Germany), formaldehyde, urea, formic acid, NH₄Cl in technical grade, and areca leaf sheaths from PT Greenie Alam Indonesia (GAI). The preparation of lignosulfonate and Eucalyptus lignin is referred to by Madyaratri et al. [14].
An ultra-low-emitting formaldehyde UF resin with a F/U molar ratio of 0.8 was prepared. Formaldehyde was placed in a beaker and heated to 40 °C and pH 8.0 while stirring with a digital stirrer at 300–350 rpm. NaOH solution was gradually added if the pH of the solution was still low. The first urea was gradually added to the mixture, and pH 8.0 was maintained until the mixture reached 90 °C after stirring for one hour. After one-hour, formic acid was gradually added to the solution to lower the pH to 4.5–5.0. The temperature was held at 80 °C for 2.5 h, with the pH of the solution reaching 4.6, at which point a second urea was added, and the mixture was stirred for 30 min. The temperature was lowered to 25 °C, followed by adding NaOH solution to reach pH 8.0. The adhesive solution was then stored in a sealed container.

The research flow chart is presented in Figure 1. Lignosulfonate was dissolved in water at 10% concentration, and lignin was dissolved at 5% concentration in NaOH solution before mixing with UF adhesive and adding 3% NH₄Cl as an adhesive hardener. The adhesive solution was mixed with areca powder in a drum mixer with a compressor and a spray gun before being poured into a 25 cm × 25 cm × 1.2 cm mold. The mixtures were heated under pressure for 10 min at 150 °C, and the resulting boards were conditioned for 7 days at room temperature (20 ± 2 °C) and 65% relative humidity. The resulting particle board has a target density of 0.8 g/cm³ and tests for determining the laboratory-fabricated boards’ physical, mechanical, and fire-resistant characteristics were conducted. A set of control boards was produced using UF adhesive alone without adding lignosulfonate or lignin. The different types of particleboards produced in this study are listed in Table 1.

![Research flow chart](image)

**Figure 1.** Research flow chart.

**Table 1.** The particleboard’s code.

<table>
<thead>
<tr>
<th>Treatment</th>
<th>Code</th>
</tr>
</thead>
<tbody>
<tr>
<td>Particleboards-control</td>
<td>PK</td>
</tr>
<tr>
<td>Particleboards bonded with UF resin and commercial lignin</td>
<td>PL</td>
</tr>
<tr>
<td>Particleboards bonded with UF resin and isolated lignin after three washings</td>
<td>PL3</td>
</tr>
<tr>
<td>Particleboards bonded with UF resin and isolated lignin after five washings</td>
<td>PL5</td>
</tr>
<tr>
<td>Particleboards bonded with UF resin and commercial lignosulfonate</td>
<td>PLs</td>
</tr>
<tr>
<td>Particleboards bonded with UF resin and lignosulfonate from isolated lignin with three washings</td>
<td>PLs3</td>
</tr>
<tr>
<td>Particleboards bonded with UF resin and lignosulfonate from isolated lignin with five washings</td>
<td>PLs5</td>
</tr>
</tbody>
</table>

2.1.1. Density

A 100 mm × 100 mm × 10 mm test sample was used for the particleboard density test. The test sample was weighed using dry air weight, and the volume was calculated using measurements of the sample’s dimensions.

\[
\text{Density (g/cm}^3\text{)} = \frac{\text{weight of sample}}{\text{volume of sample}}
\]

2.1.2. Moisture Content

The test sample from the density test was utilized again to determine the moisture content of the particleboards. The test sample was dried at 103 ± 2 °C for 24 h before being placed in a desiccator and measured to determine the oven’s dry weight.

\[
\text{Moisture content (\%)} = \left(\frac{\text{initial weight of sample} - \text{oven dry weight of sample}}{\text{oven dry weight of sample}}\right) \times 100\%
\]

2.1.3. Water Absorption

A sample of 50 mm × 50 mm × 10 mm was used for the water absorption capacity test. The test sample was initially weighed (B1), then immersed in water at room temperature for 24 h, drained, and the final weight (B2) determined.

\[
\text{Water absorption (\%)} = \left(\frac{B2 - B1}{B1}\right) \times 100\%
\]

2.1.4. Thickness Swelling

The thickness swelling test sample was utilized for the water absorption test. The thickness of the test sample was measured (t1), then subsequently measured (t2) after being immersed in water for 24 h at room temperature.

\[
\text{Thickness swelling (\%)} = \left(\frac{t2 - t1}{t1}\right) \times 100\%
\]


2.2.1. Modulus of Rupture (MOR)

The ability of interior wood materials to withstand loads is known as the modulus of rupture (MOR). A test sample measuring 20 cm × 5 cm × 1 cm was used to determine the fracture toughness using a universal testing machine (UTM).

\[
\text{MOR} = \frac{3BL}{2bh^3}
\]

where

- MOR is the modulus of rupture (kgf/cm²);
- B is the maximum load (kgf);
- L is the span length (cm);
- b is the average width of the specimen (cm);
- h is the average thickness of the specimen (cm).

2.2.2. Modulus of Elasticity (MOE)

Modulus of elasticity (MOE) testing was conducted concurrently using the same test sample as MOR testing. The deflection at a specific load period is noted when testing the MOE.

\[
\text{MOE} = \frac{\Delta P L^3}{4\Delta Y bh^3}
\]
where
- **MOE** is the modulus of elasticity (kgf/cm²);
- **ΔP** is the maximum load difference (kgf);
- **L** is the span length (cm);
- **ΔY** is the deflection that occurs in the difference in load (cm);
- **b** is the average width of the specimen (cm);
- **h** is the average thickness of the specimen (cm).

2.2.3. Internal Bond Strength (IB)

A test sample with a known surface area (A) and dimensions of 50 mm × 50 mm × 10 mm was used for determining the internal bond strength (IB). After that, epoxy adhesive was used to adhere the steel beams to the test sample, which measured 50 mm × 50 mm, and was allowed to cure. The test sample was set up on the UTM machine, and a steel beam was pushed parallel to the ground until it reached the maximum load (P).

\[
IB = \frac{P}{A}
\]

2.3. Statistical Analysis

This research was conducted using statistical analysis with a one-factor completely randomized design (CRD), namely differences in filler type (lignin and lignosulfonate) on the mechanical properties of particleboard with three replications. The data were analyzed using analysis of variance (ANOVA) to evaluate the variability of the observed data, conducted with IBM SPSS Statistics (a statistical package for service solutions) version 20.0 with a confidence level of 95%. If the results obtained were significant, Duncan's further test was conducted to determine which variables were significantly different. The linear model used was as follows:

\[
Y_{ij} = \mu + A_i + \epsilon_{ij}
\]

where
- **\(Y_{ij}\)**: Observation value at the **i**-th level in the **j**-th repetition.
- **\(\mu\)**: General mean.
- **\(A_i\)**: The effect of filler difference on the **i**-th level.
- **\(\epsilon_{ij}\)**: Effect of error on the difference factor of **i**-th filler and **j**-th repetition.
- **i**: Filler differences at control level, PK, PLs, PLs3, PLs5, PL, PL3, and PL5.
- **j**: Repetitions.

2.4. Fire Resistance Characteristics

Flammability was tested using a vertical burning test conducted in accordance with the UL−94 standard, as shown in Figure 2a (Serpong, Indonesia). The test specimens with dimensions of 125 mm × 13 mm × 12 mm were prepared from laboratory-made particleboards and bonded with an adhesive system comprising UF resin, lignin, or lignosulfonate. The sample was burned for 10 s at a predetermined distance using methane gas. The time (t) from the start of the fire until the fire was put out was then calculated. In this testing, materials were given one of three ratings: V−0 (flame extinguishes in 10 s without dripping), V−1 (flame extinguishes in 30 s without dripping), or V−2 (flame extinguishes in 10 s with dripping).

The boards that had been treated with lignosulfonate, lignin, or control were manually burned using a torch gas (Figure 2b) for three minutes in accordance with SNI1740–2008 [20] with modifications. The sample was then weighed and subjected to analysis after being oven-dried for 24 h at 105 °C. The burned sample was reweighed when the fire had completely burned out.
isolated lignin with five washings) is light blue color. PLs5 (Particleboards bonded with UF resin and lignosulfonate from isolated lignin with three washings) is green color, PLs3 (Particleboards bonded with UF resin and lignosulfonate from isolated lignin with five washings) is light blue color, PL3 (Particleboards bonded with UF resin and isolated lignin after three washings) is orange color, PL5 (Particleboards bonded with UF resin and isolated lignin after five washings) is dark purple color, PL (Particleboards bonded with UF resin and commercial lignin) is dark color, the sample after flammability test.

Figure 2. Flammability testing by UL-94 in vertical burning tests (a) and torch gas (b), burned samples.

3. Results and Discussion
3.1. Physical Characteristics

As shown in Figure 3, the moisture content of the particleboards produced in this study ranged between 9.06 and 9.61%. The overall moisture level of the areca particleboards was still within the acceptable range of JIS A 5908-2003, which is between 5 and 13 %. The moisture content of the raw material and the method of manufacture can both affect the moisture content of the finished particleboard. Before being combined with the adhesive as a chemical flame retardant, the lignosulfonate is first dissolved in distilled water, or the lignin is dissolved in a NaOH-distilled water solution. Therefore, it might be the reason that the moisture content of the particleboards made with lignin and lignosulfonate was higher than the control.

Figure 3. The moisture content of particleboards produced. (Error bars represent the standard deviation, and the horizontal line represents the JIS 5908-2003 standard requirement). PK (particleboards-control) is dark purple color, PL (Particleboards bonded with UF resin and commercial lignin) is dark blue color, PL3 (Particleboards bonded with UF resin and isolated lignin after three washings) is orange color, PL5 (Particleboards bonded with UF resin and isolated lignin after five washings) is light purple color, PLs (Particleboards bonded with UF resin and commercial lignosulfonate) is red brown color, PLs3 (Particleboards bonded with UF resin and lignosulfonate from isolated lignin with three washings) is green color, PLs5 (Particleboards bonded with UF resin and lignosulfonate from isolated lignin with five washings) is light blue color.
The densities of the control particleboard and those made with the addition of lignin or lignosulfonate solution are nearly the same, ranging from 0.79 to 0.84 g/cm³ (Figure 4). The density of all laboratory-fabricated particleboards meets the requirements of the JIS A 5908-2003 standard. The amount of adhesive used has an impact on the density value. The density value increases with the amount of adhesive used since the adhesive is distributed more evenly throughout the material, as a result of which the composite board has a greater density value [21]. Using a spray gun to mix the particles with the adhesive, on the other hand, influences how evenly the adhesive spreads out. Because of the binding between the particles and the adhesive, the particleboard’s density will increase, reducing the number of pores [22]. The inclusion of fire-retardant additives was expected to fill the board’s gaps and increase its density, but the treated board’s value was not significantly different from the density of the control boards, bonded only with UF resin. This is most likely caused by the particles’ close bindings or the additive’s size, which is larger than the size of the spaces between the particles. The particleboard density value will affect many different tests, including thickness swelling, water absorption, MOE, MOR, and IB.

![Figure 4. The density of particleboards produced. (Error bars represent the standard deviation, and the horizontal line represents the JIS 5908-2003 standard requirement). PK (particleboards-control) is dark purple color, PL (Particleboards bonded with UF resin and commercial lignin) is dark blue color, PL3 (Particleboards bonded with UF resin and isolated lignin after three washings) is orange color, PL5 (Particleboards bonded with UF resin and isolated lignin after five washings) is light purple color, PLs (Particleboards bonded with UF resin and commercial lignosulfonate) is red brown color, PLs3 (Particleboards bonded with UF resin and lignosulfonate from isolated lignin with three washings) is green color, PLs5 (Particleboards bonded with UF resin and lignosulfonate from isolated lignin with five washings) is light blue color.](image)

Water absorption and thickness swelling are important physical properties of particleboards and are strongly correlated with the dimensional stability of laboratory-produced boards [23,24]. Both properties were determined after 24 h of immersion in water.

The thickness swelling values of the particleboards after 24 h immersion in water ranged from 72% to 89%, as shown in Figure 5. The thickness swelling of all laboratory particleboards manufactured from areca leaf sheaths, bonded with an adhesive mixture of ULEF UF resin and technical lignins, exceeded the maximum allowable value as specified in JIS A 5908-2003, which is 12%. The deteriorated thickness swelling values of the laboratory boards may be explained by the exceedingly hygroscopic nature of particleboards bonded with the addition of technical lignins. More research is needed to improve the particleboard’s water resistance. In this study, it can be observed that particleboards made with the addition of lignin and lignosulfonate had lower thickness swelling values.
The water absorption capacity of the particleboards treated with lignin or lignosulfonate solution and the untreated boards varied from 129% to 160% (Figure 6). These findings are considerably different from the research of Fitra et al. [25], which indicates that particleboard constructed from areca nut shells and tapioca had thickness swelling values ranging from 51% to 168%. The raw materials employed may have an impact on this. The density of the board has an impact on the particleboard’s capacity to absorb water, with a higher density resulting in a reduced capacity [26]. The size of the particles used can have an impact on the amount of water absorption in the particle board. The smaller the particle size, the lower the water absorption because fewer spaces may be filled with water [27].
Water absorption is lower in particleboard made from a combination of lignin solutions than in lignosulfonate solutions. This could be because the lignin used was separated from black liquor using the Kraft process and has water-insoluble properties or is difficult to bind to water [14,28]. These outcomes also correspond to the characteristics of swelling at lesser thicknesses, as seen in Figure 3. Essentially, the lignosulfonates used to make boards have qualities that make them water-soluble [29]; therefore, during the immersion process, a significant amount of lignosulfonates are dissolved in water, leading to empty voids in the filled boards.

3.2. Mechanical Characteristics

With regard to the mechanical properties, the modulus of rupture (MOR), modulus of elasticity (MOE), and internal bond (IB) strength of the laboratory-fabricated particleboards were determined. The MOR values of the particleboards produced in this study are shown in Figure 7. Except for the PLs particleboard, all particleboard treatments containing lignin and lignosulfonate produced higher MOR values than the control board and met the JIS A 5908:2003 standard of 82 kg/cm². The highest MOR value was obtained in the PLs5 particleboard sample, with a value of 113.49 kg/cm², while the smallest value in the PLs particleboard sample was 77.13 kg/cm².

![Figure 7. Modulus of rupture (MOR) of particleboards produced. (Error bars represent the standard deviation, and the horizontal line represents the JIS 5908-2003 standard requirement). PK (particleboards-control) is dark purple color, PL (Particleboards bonded with UF resin and commercial lignin) is dark blue color, PL3 (Particleboards bonded with UF resin and isolated lignin after three washings) is orange color, PL5 (Particleboards bonded with UF resin and isolated lignin after five washings) is light purple color, PLs (Particleboards bonded with UF resin and commercial lignosulfonate) is red brown color, PLs3 (Particleboards bonded with UF resin and lignosulfonate from isolated lignin with three washings) is green color, PLs5 (Particleboards bonded with UF resin and lignosulfonate from isolated lignin with five washings) is light blue color.](image)

The highest MOR value of 103.60 kg/cm² was observed in the particleboard made with eucalyptus lignin after 5x washing treatments. The results obtained are lower than previous research using oil palm trunk with 10% UF, resulting in a MOR of 207.61 kg/cm² [30] and high-density fiberboard (HDF) with UF adhesive, with the addition of ALS (ammonium lignosulfonate) of as much as 4% and 6%, obtaining MOR values of 366 and 375 kg/cm² [31]. This is because adding lignin and lignosulfonate as fillers can increase particleboard density, resulting in a higher MOR value [32]. However, in the PLs treatment, the density value (0.79 g/cm³) was lower than the other treatments, resulting in a lower MOR value than the other adhesive systems.
Figure 8 depicts the MOE test results of areca leaf particleboard bonded with the addition of lignin and lignosulfonate and ULEF UF adhesive. The results showed that the highest MOE value was 10,663 kg/cm² in sample PLs5, while the lowest value was 8736 kg/cm² in sample PLs3. The highest MOE value in the lignin treatment was PL3 (lignin eucalyptus 3×) at 10,075 kg/cm². These results are lower than previous research using UF adhesives with ALS (ammonium lignosulfonate) of 4% and 6% on HDF, which has MOE values of 37,974 to 39,361 kg/cm² [33]. The addition of polymeric materials such as lignin or lignosulfonate as fillers can fill pores or cavities to bind them together [34]. Based on the test results, all treatment variations did not fulfill the JIS A 5908:2003 standard, which requires a minimum particleboard MOE value of 20,400 kg/cm². It is suspected that there are still many air voids in particleboard that are not completely filled with filler, so the bond between the matrix and filler is weakened and can result in a smaller MOE value [34,35]. The use of different types of fillers can produce different MOE values. The control particleboard obtained a higher MOE value compared to the particleboard using fillers, except for the PLs5 particleboard (Lignosulfonate Eucalyptus 5×).

![Figure 8. Modulus of elasticity (MOE) of particleboards produced. (Error bars represent the standard deviation, and the horizontal line represents the JIS 5908-2003 standard requirement). PK (particleboards-control) is dark purple color, PL (Particleboards bonded with UF resin and commercial lignin) is dark blue color, PL3 (Particleboards bonded with UF resin and isolated lignin after three washings) is orange color, PL5 (Particleboards bonded with UF resin and isolated lignin after five washings) is light purple color, PLs (Particleboards bonded with UF resin and commercial lignosulfonate) is red brown color, PLs3 (Particleboards bonded with UF resin and lignosulfonate from isolated lignin with three washings) is green color, PLs5 (Particleboards bonded with UF resin and lignosulfonate from isolated lignin with five washings) is light blue color.](image)

The IB values of particleboards varied from 5.38 to 6.96 kg/cm² (Figure 9). All particleboard treatments met the JIS A 5908:2003 standard requirement of 1.5 kg/cm². The lowest IB value in the PLs treatment of 5.38 kg/cm² was lower than the control board, while the highest IB in the PLs3 treatment was 6.96 kg/cm². In contrast, the highest IB value in the treatment of particleboard with lignin was 6.87 kg/cm² in the PL (lignin reference) treatment. The addition of lignin and lignosulfonate to the adhesive system did not result in significantly different values compared with the control panel. This is because lignin has a low reactivity to UF adhesives, so there is a decrease in bond strength [36]. Research results by Bekhta et al. [37] found that particleboard bonded with UF adhesives produced higher IB values than UF adhesives added with lignosulfonate (either magnesium lignosulfonate or sodium lignosulfonate). Whereas in other studies, using UF adhesives with the addition
of ammonium lignosulfonate at 8% and 10% resulted in higher IB strength compared to the control board [33]. Cetin and Ozmen [38] stated that the use of 20–30% organosolv lignin could replace phenol in phenol-formaldehyde (PF) adhesives, which can bind particleboard and does not affect IB strength. Small particle sizes can also cause high IB values by adding fillers that can fill empty spaces and produce large surface areas so that there is good bonding between the filler and the matrix [39]. The density of the board can influence the properties of particleboard; the denser the board, the better its mechanical properties [40].

![Figure 9. Internal bond (IB) of particleboards.](image)

According to the statistical analysis of a one-factor ANOVA on mechanical property testing, as shown in Table 2. The analysis results show that only the MOR test was significant for the treatment of areca leaf sheath particleboard, shown at $p < 0.05$. Meanwhile, MOE and IB tests were not significant. Then, further tests using Duncan’s analysis are conducted to evaluate specific differences in particleboard treatments, as shown in Table 3.

### Table 2. Statistical analysis with one-factor ANOVA on mechanical properties of particleboard.

<table>
<thead>
<tr>
<th>Source</th>
<th>Degree of Freedom</th>
<th>Sum of Squares</th>
<th>Mean Square</th>
<th>F-Value</th>
<th>p-Value *</th>
</tr>
</thead>
<tbody>
<tr>
<td>Modulus of rupture (MOR)</td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
</tr>
<tr>
<td>Between Groups</td>
<td>6</td>
<td>2933.87</td>
<td>488.98</td>
<td>3.675</td>
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<td>Within Groups</td>
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<td>1862.93</td>
<td>133.07</td>
<td></td>
<td></td>
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<tr>
<td>Total</td>
<td>20</td>
<td>4796.80</td>
<td></td>
<td></td>
<td></td>
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<tr>
<td>Modulus of elasticity (MOE)</td>
<td></td>
<td>9,227,591.78</td>
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<td>0.939</td>
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<tr>
<td>Within Groups</td>
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<td>5,553,194.99</td>
<td></td>
<td></td>
</tr>
<tr>
<td>Total</td>
<td>20</td>
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<td>Internal bond strength (IB)</td>
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<td>Within Groups</td>
<td>14</td>
<td>21.67</td>
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</table>

* $p < 0.05$.  

**Note:** Figure 9 shows the internal bond strength (IB) of particleboards produced. Error bars represent the standard deviation, and the horizontal line represents the JIS 5908-2003 standard requirement.
Table 3. Duncan’s further test analysis results.

<table>
<thead>
<tr>
<th>Treatment</th>
<th>MOR</th>
<th>MOE</th>
<th>IB</th>
</tr>
</thead>
<tbody>
<tr>
<td>PK</td>
<td>79.71 ± 7.51 c</td>
<td>10412.06 ± 3379 a</td>
<td>6.52 ± 1.41 a</td>
</tr>
<tr>
<td>PLs</td>
<td>77.13 ± 21.26 c</td>
<td>8932.42 ± 1978 a</td>
<td>5.38 ± 1.46 a</td>
</tr>
<tr>
<td>PLs3</td>
<td>95.26 ± 3.02 abc</td>
<td>8736.15 ± 2176 a</td>
<td>6.96 ± 1.19 a</td>
</tr>
<tr>
<td>PLs5</td>
<td>113.49 ± 7.80 a</td>
<td>10663.42 ± 1458 a</td>
<td>6.25 ± 1.23 a</td>
</tr>
<tr>
<td>PL</td>
<td>94.53 ± 8.73 abc</td>
<td>9816.21 ± 968 a</td>
<td>6.87 ± 0.87 a</td>
</tr>
<tr>
<td>PL3</td>
<td>89.72 ± 14.56 bc</td>
<td>10074.73 ± 3813 a</td>
<td>6.80 ± 0.26 a</td>
</tr>
<tr>
<td>PL5</td>
<td>103.6 ± 8.04 ab</td>
<td>9783.29 ± 1096 a</td>
<td>6.18 ± 0.51 a</td>
</tr>
</tbody>
</table>

Note: Different superscript letter means significant difference based on Duncan’s test at α = 0.05 and vice versa. The PK and PLs treatments were not significantly different (notation c) on MOR, while the PLs3 and PL treatments were also not significantly different (notation abc) on MOR, but the PLs5 treatment has a significant difference (notation a) with the PK and PLs treatments on MOR. The PL3 treatment (notation bc) is not significantly different from PL5 treatment (notation ab).

3.3. Fire Resistance Characteristics

The widespread usage of halogen-containing flame-retardant chemicals in composite items raises concerns about their potential health effects on people. This research aimed to use lignin-based fire-retardant compounds to enhance the fire resistance of particleboards manufactured from areca leaf sheaths as agricultural by-products. Lignin has strong thermal stability characteristics, making it a viable candidate for use as a fire-retardant substance. The particleboard’s fire-retardant characteristics are anticipated to improve by including lignin and lignosulfonate as additives. It stands to reason that using fire-resistant materials on particleboard can help contain a fire when it starts. Table 4 demonstrates no flame time after combustion, indicating that the particleboard samples have an excellent flammability rating. This could be because a raw material’s fire resistance improves after being transformed into particle board due to its higher density following adhesive application [22]. Additional testing is necessary to obtain more precise results on the fire resistance characteristics of particleboard.

Table 4. UL-94 classification of composite boards.

<table>
<thead>
<tr>
<th>Treatment</th>
<th>t after Flame (s)</th>
<th>t after Extinguished (s)</th>
<th>Dripping</th>
<th>Rating</th>
</tr>
</thead>
<tbody>
<tr>
<td>PK</td>
<td>0</td>
<td>0</td>
<td>No</td>
<td>V–0</td>
</tr>
<tr>
<td>PL</td>
<td>0</td>
<td>0</td>
<td>No</td>
<td>V–0</td>
</tr>
<tr>
<td>PL3</td>
<td>0</td>
<td>0</td>
<td>No</td>
<td>V–0</td>
</tr>
<tr>
<td>PL5</td>
<td>0</td>
<td>0</td>
<td>No</td>
<td>V–0</td>
</tr>
<tr>
<td>PLs</td>
<td>0</td>
<td>0</td>
<td>No</td>
<td>V–0</td>
</tr>
<tr>
<td>PLs3</td>
<td>0</td>
<td>0</td>
<td>No</td>
<td>V–0</td>
</tr>
<tr>
<td>PLs5</td>
<td>0</td>
<td>0</td>
<td>No</td>
<td>V–0</td>
</tr>
</tbody>
</table>

Three minutes of burning a gas torch on a particle board resulted in varying degrees of weight loss (Table 5). The least amount of weight was lost when lignosulfonate (PLs5) was applied to the particleboard as a fire retardant. Lignosulfonates are more fire-resistant and thermally stable than lignin, according to research by Madyaratri et al. [14]. These FR additives’ flame-retardant properties depend on the solid phase’s ability to cool down by releasing water molecules while also encouraging the creation of a protective layer [41]. The extent to which fire-resistant materials are used, the portion of the burned biomass, and the charring process all affect how long the combustion process lasts in biomass [42]. The inclusion of flame-retardant components, which served as a fire barrier and aided in the formation of a charred structure, exhibited favorable fire-protective characteristics. This might occur as a result of chemical or physical reactions between the adhesive and the flame-retardant components [43].
Table 5. Flammability test of composite boards with gas torch for 3 min.

<table>
<thead>
<tr>
<th>Treatment</th>
<th>Weight before Flame (g)</th>
<th>Weight after Flame (g)</th>
<th>Mass Loss (%)</th>
</tr>
</thead>
<tbody>
<tr>
<td>PK</td>
<td>15.99</td>
<td>11.29</td>
<td>29.39</td>
</tr>
<tr>
<td>PL</td>
<td>15.57</td>
<td>5.88</td>
<td>62.24</td>
</tr>
<tr>
<td>PL3</td>
<td>15.78</td>
<td>4.52</td>
<td>71.36</td>
</tr>
<tr>
<td>PL5</td>
<td>16.1</td>
<td>4.5</td>
<td>72.05</td>
</tr>
<tr>
<td>PLs</td>
<td>16.5</td>
<td>8.23</td>
<td>50.12</td>
</tr>
<tr>
<td>PLs3</td>
<td>17.62</td>
<td>12.38</td>
<td>29.74</td>
</tr>
<tr>
<td>PLs5</td>
<td>17.55</td>
<td>14.62</td>
<td>16.70</td>
</tr>
</tbody>
</table>

4. Conclusions

This study demonstrated that environmentally friendly particleboards with acceptable physical and mechanical characteristics and improved fire resistance could be produced from areca leaf sheaths, bonded with an adhesive system comprised of ULEF UF resin and technical lignins (kraft lignin or lignosulfonate) as bio-based additives.

1. The particleboards with lignin solution had the highest density of 0.84 g/cm$^3$ (PL3), the lowest moisture content, thickness swelling, and water absorption of 9.35% (PL3), 72.16% (PL), and 131.65% (PL), and the highest MOE, MOR, and IB at 987.99 kg/cm$^2$ (PL3), 10.16 kg/cm$^2$ (PLSs), and 0.67 kg/cm$^2$ (PLs).

2. The particleboards with lignosulfonate solution had the highest density at 0.83 g/cm$^3$ (PLs3), the lowest moisture content, thickness swelling, and water absorption at 9.06% (PLs3), 83.61% (PLs5), and 129.17% (PLs5), and the highest MOE, MOR, and IB at 1045.72 kg/cm$^2$ (PLs5), 11.13 kg/cm$^2$ (PLs5), and 0.68 kg/cm$^2$ (PLs).

3. All the particleboards had a rating of V-0 for the UL-94 test, and the lowest mass loss for the torch gas test was 16.70% (PLs5). The weight loss of particleboard can be minimized by adding additives with lignosulfonate as fire retardants.

Future studies should focus on modifying the lignin additives to improve their reactivity towards formaldehyde and thoroughly investigating the bonding processes between the UF resin, lignin additives, and lignocellulosic particles.

5. Patents

The Indonesian Patent Officer has registered this invention with the patent registration number P00202304423.


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Conflicts of Interest: The authors declare no conflict of interest.

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