ORIGINAL RESEARCH

UTILIZATION OF LIGNIN ISOLATION RESULTS FROM OIL PALM FRONDS AS ADHESIVE IN COMPOSITE BOARD PREPARATION

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Abstract

The decreasing availability of adhesive raw materials and the emergence of formaldehyde emissions from gluing material products to the environment, require a substitute material in the manufacture. Lignin from oil palm fronds has the potential as an alternative environmentally friendly adhesive in the composite board industry and offers a sustainable agricultural waste management solution because of its abundant availability. This study is a laboratory experiment, the utilization of lignin from oil palm fronds, which will later be applied as a composite wood adhesive. From the results of the study obtained. copolymerization with resorcinol and formaldehyde produces lignin resorcinol formaldehyde (LRF) resin which is tested for adhesive appearance, pH/acidity, specific gravity, and viscosity of LRF adhesive at room temperature and residual evaporation levels at 105 oC. Product characterization includes testing the physical and mechanical properties of composite boards including Fracture Strength (Modulus of Rufture) and Flexural Strength or Modulus of Elasticity (MOE) from the test results of this lignin-based adhesive which meets the specified requirements.

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1. Introduction

Indonesia, as a major producer of palm oil, faces challenges in the management of agricultural waste, especially palm fronds, which amount to millions of tons per year. This accumulation of waste not only poses an environmental problem but also represents a potential resource that has not been optimally utilized. One innovative approach is the utilization of lignin, a major component of plant cell walls, which can be extracted from oil palm fronds. Lignin has potential as a natural adhesive material in the composite board industry, offering an environmentally friendly alternative to synthetic adhesives that often contain harmful volatile organic compounds (VOCs) [1]. Therefore, this study aims to explore the isolation of lignin from oil palm fronds and evaluate its potential as an adhesive material in the manufacture of composite boards, in the hope of reducing environmental impacts and increasing the added value of local resource-based products [2].

The increase in palm oil production in Indonesia has led to a significant accumulation of palm frond waste. This waste is often not optimally utilized, posing challenges in waste management and potential environmental impacts. One approach to address this issue is to extract lignin from palm fronds. Lignin, as a major component of plant cell walls, has aromatic functional groups that allow interaction with other polymer matrices, making it suitable for improving the mechanical properties of composite boards [3].

Several studies have explored the use of lignin as a natural adhesive material in composite board manufacturing. For example, research by [4] showed that the composition variation between lontar fiber and epoxy resin affects the density and mechanical properties of the resulting composite board. In addition, research by [2] used the results of lignin isolation of formaldehyde resorcinol from sodium lignosulfonate of oil palm empty fruit bunches for making adhesives by reacting lignin from the purification process of sodium lignosulfonate added with formaldehyde. Research by [5] showed that lignin can function as a natural adhesive in particleboard, affecting the physical and mechanical properties of the final product. In addition, research by [6] showed that the use of lignin and tannin formaldehyde as an adhesive can improve the quality of the hardboard produced [8] using lignin from nipah fruit peel black leachate as a lignin resorcinol formaldehyde adhesive. The utilization of lignin as a wood adhesive is also carried out [8][9]which utilizes lignin from the by-products of the wood burial process to obtain resol resin by demethylating lignin and reacting demethylated lignin with aldehydes (formaldehyde, para formaldehyde or furfural) in alkaline media used for composite wood adhesives.

However, there are limitations in previous studies related to the lignin extraction method used and variations in lignin concentration as an adhesive. Therefore, this study aims to isolate lignin from oil palm fronds using an efficient method, evaluate the potential of lignin as an adhesive material in the manufacture of composite boards, and analyze the physical and mechanical characteristics of the resulting composite boards. Hopefully, the results of this research can make a significant contribution to the management of palm oil waste and the development of environmentally friendly composite materials [10].

2. Methods

Tools

The tools used in this study were pH meter, glassware, analytical scales, incubator, autoclave, oven, belender, water bath, ose needle, scaled ruler, push term and mesh sieve 200.

Materials

The materials used were Kersen leaves, Staphyloccocus aureus bacterial isolates, olive oil, potassium hydroxide (KOH), carboxyl methyl cellulose (CMC), sodium lauryl sulfate (SLS), stearic acid, butyl hydroxy toluene (BHT), 96% ethanol, Nutrient agar, detol soap, NaCl 0.9%, aluminum foil.

Research Procedures

1. **Preparation of Adhesive and Analysis of Lignin Resorcinol Formaldehyde (LRF) Adhesive**

Weighed as much as 5 grams of lignin isolation results from oil palm fronds, suspended with $NaHSO_3$ with a mole ratio of lignin: $NaHSO_3$ is 1:0.5 and then put in 150 ml of water. The lignin mixture was put into a three-neck flask with a volume of 500 mL using a stirrer. pH was set to 7 by adding 15% NaOH in the manner shown on the universal pH indicator scale. so that the mixture reacted completely the mixture was stirred using a stirrer. The mixture was then refluxed at 106 ^oC, so that the mixture reacted completely, stirring was carried out with a stirrer and electric heater for 4 hours..

The reflux results in the manufacture of sodium lignosulfonate are distilled at 100^oC to evaporate water to reduce the volume and to purify Sodium Lignosulfonate,. To separate the remaining lignin, the concentrated solution was put into a separating funnel. The filtrate is sodium lignosulfonate which still contains $NaHSO_3$ (reaction residue). The filtrate was then added to methanol while shaking vigorously so that $NaHSO_3$ precipitated and put in a separatory funnel. The filtrate was evaporated to separate the methanol contained in the filtrate. The concentrated NaLS obtained was dried in an oven at 60 ^oC.

To prepare LRF adhesive, 20 grams of sodium lignosulfonate (NaLS) was weighed and then mixed with l0% NaOH in a glass cup and stirred at room temperature. The mixture was added with 50% NaOH solution while stirring so that the pH reached 10. Then resorcinol was added little by little to the solution formed and stirred until homogeneous. The solution was conditioned until the pH reached 11 by adding 50% NaOH solution. Formaldehyde 37% solution was added while stirring. The l0% NaOH solution was added, and the mixture was stirred again until the pH of the solution reached pH 11 [11]. The resulting adhesive will be analyzed for physical properties including the appearance of the adhesive, pH/acidity, specific gravity of LRF adhesive at room temperature and residual evaporation content at 105 ^oC.

2. **Preparation and Characterization of Composite Wood Products**

Composite wood products are made on a laboratory scale using Lamina wood attached to one another with a size of 30 cm x 30 cm x 4.5 mm, each using lignin resorcinol formaldehyde (LRF) adhesive. The mixture is then molded and pressed using a hydraulic hot press at a certain temperature and pressure to form a composite board, forged for 15 minutes at 135°C. Composite wood is tested by giving a tensile load to the wood lamina that has been glued together until it is detached, carried out several times to find out constant results or take the average value of the experiment. The composite wood products obtained are tested for mechanical properties such as fracture toughness (Modulus Of Rufture), Flexural Strength or Modulus Of Elasticity (MOE), and Internal Bonding Firmness.

3. Results

Lignin Resorcinol Formaldehyde Adhesive Test Results

Isolation of lignin from palm frond powder has been successfully carried out by delignification process using NaOH cooking solution. The resulting LRF adhesive can be seen in Figure 1 below: 3

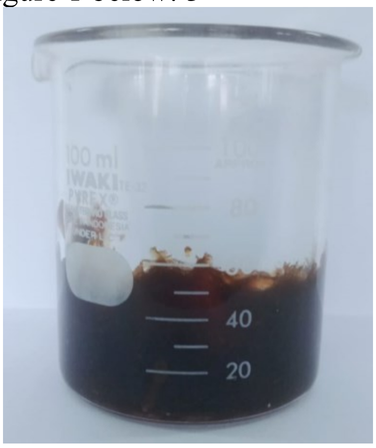


Figure 1 Lignin Resorcinol Formaldehyde (LRF) Adhesive

Lignin Resorcinol Formaldehyde can be used as an adhesive because of its ability to bind wood cells to each other, making them rigid and strong, and giving wood the strength to absorb the forces imposed on it [12].

1. Appearance Test

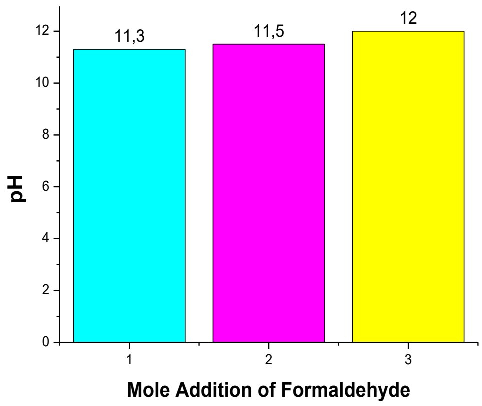
Testing the appearance of the adhesive is very important, because in this test will be known whether or not there are foreign objects contained in the adhesive in the form of dust and small granules in small quantities. The presence of foreign matter, dust and grains in the adhesive will reduce the quality of the adhesive, in Figure 1 The results of Lignin Resorcinol Formaldehyde (LRF) adhesive obtained is red brown-black and free of impurities according to SNI 06-4567-1998 quality standards of phenol formaldehyde adhesive is a liquid and its appearance is red-black and free of impurities.

According to [13] phenolic compounds contain 1 additional hydroxy group in the aromatic core forming a meta position resulting in a color derived from the combination of lignin isolate with resorcinol.

The high solids content of the adhesive was influenced by the addition of resorcinol. Thus, resorcinol influences the solids content which indicates the formation of components with larger molecules such as jelly [14] [15].

2. **pH Test (Degree of Acidity)**

The results of the pH (Degree of Acidity) test of LRF adhesive can be seen in Figure 2 below: The pH (Degree of Acidity) of LRF adhesive can be seen in



Figure

Figure 2 The effect of formaldehyde mole addition on the pH value of LRF adhesive

The pH test based on the figure above the average pH value of each comparison is 1:1:1 ratio is 11.3, 1:1:2 ratio is 11,.5 and 1:1:3 ratio is 12.0. for the addition of formaldeid has no effect on the manufacture of LRF adhesives. Because the final pH expected in the manufacture of adhesives is at pH 11, based on the pH test obtained in accordance with SNI 06-4567-1998 pH value of 10.0 - 13.0 (Rahmawati et al., 2024). The addition of 50% NaOH into the adhesive solution makes the adhesive alkaline, The alkaline nature of the adhesive is needed so that the wood structure is not damaged, because the acidic pH of 3.5 - 4 makes the adhesive strength of the wood will begin to lose its adhesive bond [15][16].

3. **Specific gravity test**

The results of the LRF adhesive specific gravity test can be seen in Figure 3 below: Specific gravity test of LRF adhesive can be seen in Figure 3 below:

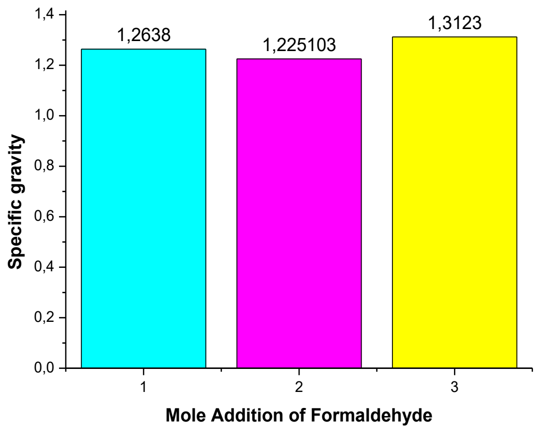


Figure 3 Effect of Formaldehyde Mole Addition on Specific Weight of LRFAdhesive

Based on the picture above, the average specific gravity value of each comparison is 1:1:1 ratio of 1.2638, 1:1:2 ratio of 1.2251 and 1:1:3 ratio of 1.3123. The resulting adhesive shows the effect of the addition of formaldehyde on the adhesive product produced.

The data in the figure above shows the specific gravity value for all comparisons is not appropriate but close to the standard value of SNI 06-4567-1998 requires a specific gravity value of 1.165 - 1.200.

The constituent components of the adhesive affect the specific gravity of the adhesive, where the more constituent components of the adhesive will result in an increased specific gravity value [17].

4. Residual Evaporation Test

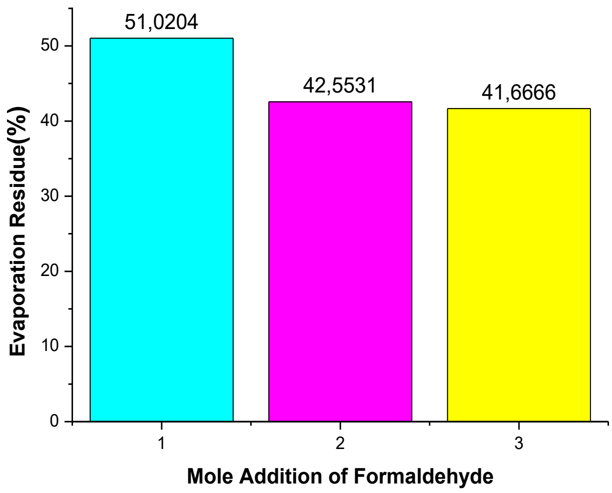


Figure 4 Effect of Formaldehyde Mole Addition on Residual Evaporation

Based on the figure above, the average residual evaporation value of LRF adhesive type from each comparison is 1:1:1 ratio of 51.0204, 1:1:2 ratio of 42.5531 and 1:1:3 ratio of 41.6666. The effect of the addition of formaldehyde on the adhesive product produced. Based on the data above, the residual value of evaporation in the 1:1:1 comparison is not in accordance with the standard, while for the 1:1:2 comparison and the 1:1:3 comparison are in accordance with the standard. According to the adhesive standard of SNI 06-4567-1998 the value of residual evaporation is 40-45%. From the data obtained with the addition of formaldehyde, the evaporation of LRF adhesive is reduced. The copolymerization reaction is perfect so as to increase the adhesion to be more because the solid molecules increase [18].

3.1. Observation of Physical Properties of Composite Wood

The LRF adhesive made was tested for fracture strength and adhesive strength on composite boards made on a laboratory scale in the form of wood lamina attached to one another with a size of 30 cm x 30 cm x 4.5 mm, each using lignin resorcinol formaldehyde (LRF) and forged at 135oC for 15 minutes. After that, it was tested by giving a tensile load on the wood lamina that had been glued until it was released, carried out several times to find out the constant results or take the average value of the experiment whose results are presented in tables 1 and 2 below:

Table 1 Adhesive quality of composite wood products using LRF adhesive

LRF Resin Experiments	Products	Adhesive Firmness (Kg/cm^2)	
		Dry Test	**Wet Test**
1	Wood Lamina	74.52 – 78.11	37.22 – 43.71
2	Wood Lamina	65.10 – 70.98	42.50 – 46.12
3	Wood Lamina	68.87 – 86.92	43.79 – 45.76

Table 2 MOE and MOR values of composite wood products using LRF adhesive

Waktu kempa (menit) pada Temperatur 135^{o}C	(Kg/cm^2)	
	MOE	**MOR**
10	48.134.54	382.75
15	70.532.34	482.65
20	61.654.21	665.74

Because the adhesive firmness value of plywood using LRF adhesive is more than 7 kg/cm2, the requirement (SNI 01-50082, 2000) for exterior type is met. The results of this study are higher than similar products using commercial PF adhesives [19].

The use of LRF adhesives (1-3) on three types of wood lamina produces adhesive strength that meets the requirements of Japanese standards (JAS, 2003) because each value is > 54 kg/cm2 (dry test) and > 41 kg/cm2 (wet test) so that it can be applied to parquet flooring. This lignin-based adhesive in addition to exterior quality, is also environmentally friendly and more economical compared to phenol formaldehyde, resorcinol formal-dehyde, and phenol resorcinol formaldehyde adhesives [2].

4. Conclusion

The results of the research conducted showed that lignin isolated from oil palm fronds can serve as a natural adhesive material in the manufacture of composite boards. The resulting composite board has copolymerized with resorcinol and formaldehyde to produce resorcinol formaldehyde (LRF) lignin resin which was tested for appearance of blackish red adhesive and free of impurities, pH/acidity of 11.3, 11.5 and 12.0, specific gravity of 1.2638, 1.2251 and 1.3123, and residual evaporation content at 105 ^{o}C of 51.0204%, 42.5531% and 41.6666%.

Testing the physical and mechanical properties of composite boards including Fracture toughness (Modulus of Rufture) of 382.75 Kg/cm2, 482.65 Kg/cm2 and 665.74 Kg/cm2, Flexural Strength or Modulus of Elasticity (MOE) of 48,134.54 Kg/cm2, 70,532. 34 Kg/cm2 and 61,654.21 Kg/cm2 and Dry adhesive strength of 74.52 - 78.11 Kg/cm2, 65.10 - 70.98 Kg/cm2 and 68.87 - 86.92 Kg/cm2 Wet adhesive strength of 37.22 - 43.71 Kg/cm2, 42.50 - 46.12 Kg/cm2 and 43.79 - 45.76 Kg/cm2. The adhesive based on lignin fulfills the requirements that have been set.

This research not only offers an alternative to environmentally friendly adhesives but also increases the added value of agricultural waste, especially palm fronds.

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