Production and Properties of Liquefied Oil-palm Stem Adhesive

Arif Nuryawan[1], Iwan Risnasari[1], Hana Pratiwi Sihombing[2] and Diana Situmorang[2]
[1]*Department of Forest Products Technology, Faculty of Forestry, Universitas Sumatera Utara, Medan, Indonesia*
[2]*Faculty of Forestry, Universitas Sumatera Utara, Medan, Indonesia*

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Abstract: The objective of this study was to evaluate production and properties of liquefied oil-palm stem adhesive. Method for production of this adhesive was inspired by bio-refinery concept. In this context, vascular bundle of oil-palm stem was liquefied through liquefaction process involving thermo-chemical reaction using chemicals of H_2SO_4, phenol, and NaOH at 90°C. Properties of the adhesive were compared to Indonesian Standard for phenol-formaldehyde (PF) resins. Results of the examinations showed color of the liquefied adhesive was brown-blackish, specific gravity was 1.26, solid content was 86%, viscosity was 111 cP, pH was 7.8, gel time was 98 minutes, and free formaldehyde was not detected. Even though the properties of the liquefied adhesive made of vascular bundles' oil-palm stem were slightly differ from PF resin, application of this adhesive as binder of wood product should be recommended because of its feasibility as the adhesive.

1 INTRODUCTION

Plantation forest and oil-palm plantation have been expand since the existence of natural forest become decreasing. Plantation forest generates wood, not only from main log but also from thinning and pruning volumes, which be used for wood industry's raw material such as moulding wood, core-plywood, particleboard, fibreboard, pulp and paper. Even though oil-palm plantation mainly produces fruits for manufacturing palm oil for cooking purpose, huge amount of biomass is also being generates regularly, such as leaves, frond, empty fruit bunch, and trunk. These lignocelluloses materials up till now have been still limited in utilization.

It was known that oil-palm leaves have been used for decades only as cattle animal feed. Advanced processing of these materials has been recommended for health and disease prevention for humans (Mohamed, 2014) including for their skin caring (Yusof et al., 2016). Related to product substitution of wood, Nuryawan & Rahmawaty (2018) reported that particleboards made of oil-palm leaves have unsatisfying physical and mechanical properties therefore more treatments should be applied such as addition more hardener, application of surface layer, or replacement interior type adhesive into exterior's one.

Both frond and empty fruit bunch were also still limited in utilization. The first was mainly used as roughage source in pellet for ruminant feeding (Zahari et al. 2002) and the latter has been started to be utilized as bio-refinery feedstock (Tan et al., 2016) to produce fine chemicals, bio-fuel, and green polymeric materials.

Similarly, oil-palm trunk tends to be converted into advanced materials via bio-refinery concept rather than conventional one. Since its properties are very different from wood, utilization in solid form is less promising. It is very hygroscopic in the nature; consequently, it raises dimensional instability thus less durable. Many attempts have been done for improving these unbeneficial properties by employing whole stem into products, for example laminated combination using tropical wood (Nordin et al., 2004), engineered composite board (Wahab et al., 2013), or resin impregnated to the stem (Rosli et al., 2016).

Anatomically, oil-palm stem comprised of vascular bundles embedded in parenchyma tissues (Lim & Gan, 2005). The presence both of these parts influences the characteristics further (Ramle et al., 2012), hence some scientists separated between the two prior to further processing and utilized only one part, for instances parenchyma tissue for obtaining sugar (glucose and xylose) (Mansor & Ahmad,

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1990) and producing starch for making wood adhesive (Salleh et al., 2015) even though the yield was very limited and depended on position on the stem; bottom, middle, or upper part. Conversely only limited reports employed vascular bundles for raw materials, such as making composite plastics using matrix of polyethylene (Lubis et al., 2009) and producing oriented strand board (OSB) in laboratory scale (Nuryawan, 2010).

Therefore in this research, utilization of vascular bundles obtained from oil-palm stem was investigated which was inspired by bio-refinery concept. In this context, vascular bundle was liquefied through liquefaction process involving thermo-chemical reaction and resulting in various bio-copolymers such as coatings, various polymers, carbon fibres, foams, including adhesives (Ugovšek et al., 2011). Liquefaction is one method for converting whole biomass into liquids (Jiang et al. 2018). This work is reasonable because vascular bundle is one of lignocelluloses material having nature equal to wood with lignin content up to 22 % (Chew & Bhatia, 2008; Nuryawan et al., 2012). In addition, in the recent publication (Jiang et al., 2018), both virgin and waste lignocelluloses materials have been succeed to be turned into wood adhesives.

Because this work is the initial report on utilization of vascular bundles as raw material for making wood adhesives based on bio-refinery concept, this study then discussed the preparation, processing as well as the characteristics of resulting liquefied adhesive.

2 MATERIALS AND METHODS

2.1 Preparation of the Materials

It was reported that machining and sawing properties of oil-palm stem was difficult and the quality of finished material was rough because of presence of silica (Lim & Gan, 2005). Therefore, in this experiment converting oil-palm stem was carried out using planer machine. This machine was capable to alter solid form into particle size in short time. Resulted particles then were immersed in the water for 8 hours for separating between vascular bundles and parenchyma. The vascular bundle will be remained in the sieve as residue and the parenchyma will be passed the sieve as filtrate. Resulted vascular bundles were air-dried, grinded into 20-60 mesh in size and oven-dried (103+2)°C for 24 hours.

2.2 Production of liquefied adhesive

100 g oven-dry vascular bundle with moisture content (MC) 5% was placed into beaker glass. 25 ml H_2SO_4 98% (5% phenol weight) was added and stirred gently for 30 minutes. The mixture then was conditioned in sealed beaker glass. After 24 hours, 500 ml liquid phenol (hereafter melted in 60°C) was added into the mixture and then stirred gently until appear homogenous. Sodium hydroxide (NaOH) 40% was added until the pH 8, then formalin 37% was added with ratio phenol and formaldehyde of 1:1.2. The mixture then was filtered using filter paper and the extract was heated at 90°C in the water bath for 2 hours. The extract then was kept in glass bottle prior to applying.

2.3 Determination of Basic Properties of the Liquefied Adhesive

Determination and evaluation of basic properties of resulted liquefied adhesive were carried out according to Indonesian Standard SNI 06-4567-1998 for phenol-formaldehyde (PF) resin. The properties consisted of performance, specific gravity, solid content, viscosity, pH, gel time, and free formaldehyde.

Performance of liquefied adhesive was observed either using naked eye or microscope, consisted of clarity, color, and presence of dust. Clarity of the adhesive was performed by visual observation either using naked eye or microscope. Sample of the adhesive was poured onto a petri dish or a glass slide in order to form film layer. Observation of color and presence of peculiar objects such as granules or dust particles was carried out accurately using certain magnification.

Specific gravity (*SG*) was measured using gravimetric method using picnometer and analytical balance. An empty and oven-dry picnometer was weighed (*W1*), distilled water-filled picnometer was weight (*W2*), and dextrin's adhesive-filled picnometer was also weighed (*W3*). The specific gravity was determined as in Equation (1).

$$SG = \frac{(W3-W1)}{(W2-W1} \quad (1)$$

Solid content (SC) was measured using gravimetric method using ceramic crucibles, convection oven and analytical balance. Weigh of initial sample in the crucible about 2 grams (W1) was dried in the oven at (103±2)°C for 24 h until constant weight (W2). The solid content was determined as in Equation (2).

$$SC = \frac{(W2)}{(W1)}\ x\ 100\% \quad (2)$$

Viscosity was measured using a viscometer with appropriate spindle. Sample of liquefied adhesive was placed in a 100 ml beaker glass, and measurement was conducted in room temperature (25°C) using spindle with velocity in rpm (rotary per minute).

Electronic pH meter was used to determine the acidity of liquefied adhesive sample. Sample was placed in a 100 ml beaker glass, and measurement was conducted when the sensitive electrode was soaked into the sample.

Gel time is defined as a period for gelatinization or altering liquid adhesive into solid phase. In other word, gel time is time needed for pre-polymer liquefied adhesive becomes solidifying or curing. In some cases, *i.e* formaldehyde based adhesive, determination of gel time uses aid of hardener or catalyst such as ammonium chloride (NH_4Cl) for urea-formaldehyde (UF) resin and NaOH for PF in order to make curing. In this experiment, liquefied adhesive does not require specific condition like acidic or alkaline. Therefore simple method for determining gel time was applied. About 10 grams liquefied adhesive was placed in the reaction tube, then subsequently it was positioned under 2 cm in a boiling water bath. Calculation of time was finished when the sample in the reaction tube was altered into hardened.

Free formaldehyde was determined by weighing adhesive sample 20 g (W). The sample then was mixed with 50 ml distilled water in the Erlenmeyer. Both indicators of methyl red and blue were added 2-3 drops into the mixture and subsequently it was neutralized using either hydrochloric acid (0.1N HCl) or sodium hydroxide (1N NaOH). Afterwards the mixture was added 10 ml NH_4OH 10 wt% and 10 ml NaOH. Erlenmeyer then was covered, shake, and placed in water bath at 30°C for 30 minutes. Titration using HCl was done until the colour altered from green into blue-grey and then red-purple (V_1). Using the same procedure, blank solution was made without addition of adhesive sample (V_2). Free formaldehyde (FF) then was calculated using Equation (3).

$$FF = \frac{(V1-V2)x\ 30.03}{(W)}\ x\ 100\% \quad (3)$$

3 RESULTS AND DISCUSSIONS

Production of liquefied adhesive made of oven dry particles (MC 5%) of vascular bundles of oil-palm stem without considering position on the stem resulted yield 21% with very high viscosity. This value was lower compare to previous work of Esteves et al. (2019) who made liquefied adhesive using bark and branches of eucalyptus tree. They have been obtained yield of 62% and 48%, respectively. This was probably because of different solvent used. In converting parts of eucalyptus, mixture of glycerol and ethylene glycol was used as solvent while in this work phenol was used.

This type of adhesive is suitable only for making bond-line by spreading technique such as for plywood making. However, for spraying application likewise for particleboard or fibreboard production, it is impossible. The viscosity was too high thus the adhesive could not through-out nozzle of spray gun. Therefore, solvent for lowering the viscosity was needed. In this study, we used commercial thinner for lowering the viscosity of liquefied adhesive since water as polar solvent was not capable to dissolve the liquefied adhesive.

Basic properties of this adhesive was summarized in Table 1 as follow:

Table 1. Properties of Liquefied Adhesive Made of Oil-Palm Stem

Properties	Result	Standard
Color	Brown-blackish	Red-blackish
Specific gravity	1.26	1.165-1.200
Solid content (%)	86	40-45
Viscosity (cP)	111	130-300
pH	7.8	10-13
Gel time (minutes)	98	>30
Free formaldehyde (%)	Not detected	<1

3.1 Performance

According to Indonesian Standard SNI 06-4567-1998 for PF adhesive, the colour should be red-blackish but resulted liquefied adhesive showed brown-blackish. Either peculiar granule or dust has not been found in this adhesive. The brown colour indeed originated from lignin content from oil-palm particle. Works of Risnasari & Ruhendi (2006) and

Widiyanto (2011) in making liquefied adhesive made from wood (teak, keruing and agathis) and mixture of rubber wood and bamboo, respectively, resulted in black colour. Lignin content in wood may vary among species of woody plants, some of them could reach 30% (Campbell & Sederoff, 1996) but only around 20% within vascular bundles of oil-palm stem (Nuryawan et al., 2012).

3.2 Specific Gravity

Specific gravity (SG) of resulted liquefied adhesive was 1.26. This value was little out of range within the standard (1.165-1.200) although previous works on liquefied adhesive exhibited either higher or lower values, for instances 1.23-1.25 (Risnasari & Ruhendi, 2006) and 1.153 (Widiyanto, 2011) depended on the raw materials used. Higher value of SG was more than one (>1) means the liquefied adhesive sinks down within water.

3.3 Solid Content

Solid content was percent solid after evaporation of liquid and volatile materials. Liquefied adhesive in this work showed 86%. This value was higher than that of standard with required only 40-45%. However, comparing with other adhesives such as isocyanate, this value was still lower. Isocyanate has 98-99% solid and indeed it derived from petroleum based (Nuryawan & Alamsyah, 2019).

3.4 Viscosity

It was impossible for measuring viscosity of this liquefied adhesive because of too viscous. Therefore an attempt for lowering the viscosity by mixing with commercial thinner was carried out with ratio of liquefied adhesive and thinner about 1:0.8. Generally, application of solvent for making liquefied adhesive, so called solvolytic, has been done in the early stage of liquefaction. But in this work, both in the early and final stages, addition of solvent was carried out. The final viscosity of liquefied adhesive was 111.23 cP close to the range of standard which required 130-300 cP. This viscosity enabled spraying application for producing particleboard or fiberboard.

3.5 pH

The acidity of liquefied adhesive tends to alkaline about 7.8. This condition was influenced by addition of NaOH at the final stage of liquefaction. This condition was also beneficial to substrate or adherent for surface cleaning (via dissolve the existing contaminant) and to wood structure for swelling; thus it enhanced the penetration of adhesive used (Risnasari & Ruhendi, 2006).

3.6 Gel Time

As the gel time increase, the life use of the adhesive is longer. Liquefied adhesive in this experiment needed 98 minutes for hardening or curing. This value is in accordance with the standard which required more than 30 minutes. Addition of liquefied adhesive into commercial resin adhesive lengthen the gel time (Kunaver et al., 2010).

3.7 Free Formaldehyde

In this experiment, free formaldehyde was not detected. After HCl titration, there was no colour alter which indicated there was no free formaldehyde.

4 CONCLUSIONS

Liquefied adhesive made of vascular bundle of oil-palm stem has been successfully synthesis. Properties of this adhesive generally fulfilled the standard excluding the viscosity. This study was the first attempt lowering viscosity after synthesis by mixed with commercial solvent (thinner) for proper spraying in order to produce particleboard or fibreboard.

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