Starch Based Adhesives Made From Durian Seed Through Dextrinization

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Abstract. In order to utilize durian seeds waste, production of wood adhesive made from them was carried out. Method of dextrinization was applied and characterizations of the adhesive’s properties were evaluated, namely specific gravity, viscosity, gel time, pH, clarity, color, and microscopic description including the particle’s size of the adhesives. The properties were compared to other starch based adhesives, such as sago and tapioca. For further utilization as wood adhesive, application for particleboard binder was investigated. Parameter of internal bonding of the board then was evaluated. Result of this work showed fundamental properties of dextrin adhesive made of durian seed were similar with sago and tapioca’s dextrin excluding the colour. Values of internal bonding of particleboard were varied that indicated many factors affected the quality of particleboard for instance the distribution of the adhesive.

1. Introduction
Efforts to explore new type of wood binders as well as new adhesive systems have been carried out by some scientists [1,2] because the needs of these adhesives tend to increase with increasing demand of wood products and their development. It is well-known fact that formaldehyde based resins such as urea-formaldehyde (UF), melamine-formaldehyde (MF), and phenol-formaldehyde (PF) are the most widely used adhesive in the world because of their benefits such as low production budgeting, large scale manufacturing, ease to handling, and fast curing. However these adhesives are classified as formaldehyde emitters. Formaldehyde emissions (FE) are released during the synthesis and the use of bonded products. International for Research and Cancer even stated that FE is one of sources of carcinogenic agent and it is harmful for human health [3].

Therefore, in order to substitute these formaldehyde based resins as well as environment awareness, bio-adhesives have been re-used nowadays even though their strength and durability are limited [4-6]. Bio-based adhesives definitely present advantages in terms of health and environmental issues because the parent of these adhesives was natural renewable resources (such as animals and botanical plants) [7]. Despite the raw material of bio-adhesive is mainly for human food
consumption, there are some available sources to be utilized as feed stock of bio-adhesive like waste of skin, bone, fish, blood (from animals) and residue of bark, rind and seed (from plants) [8].

Seed of durian (Durio zibethinus) is one of the underutilized residues after consumption of the flesh of fruit. Even though this seed generates high starch compare to sweet potato and cassava [9], utilization of this residue as human food is not acceptable because of ethical reason. When enjoying durian, seeds are usually exposed to the mouth. As a result, utilization of these seeds is suitable for non-human food purpose i.e. for making adhesive. Further processing by extracting the seed into starch and converting it into dextrin will enhance its functional properties thus its application in industry will expand as well. The intention features includes solubility in cold water, less moisture/water uptake, soften in its texture, and strength in adhesion [10].

Potency of these seeds is promising. Total weight of durian fruit consists of three parts, namely 60-75% rind; 20-35% flesh of fruit; and 20-35% seed [11]. North Sumatra contributes almost 20% of total production of durian fruits in Indonesia and the yield is the highest among the provinces [11]. Further, durian is a very popular fruit in South East Asia, particularly in Thailand, Philippine, Malaysia including Indonesia [12] hence the feed stock is sustainable.

Thus, the main objective of this study was to evaluate basic characteristics of dextrin made of durian seed consisted of performance/ color, pH, specific gravity, gel time, solid content, viscosity, microscopic size, and bonding feasibility. For comparison, characteristics of two commercials dextrin originated from sago and cassava were presented except for bonding feasibility. It was only done for durian seed’s dextrin and the result was compared to standard.

2. Materials and methods

2.1. Preparation of raw materials

The durian seed was collected from Medan, North Sumatra, Indonesia. Both commercial dextrin made of sago and cassava were donated by Laboratory of Food Technology, Faculty of Agriculture, Universitas Sumatera Utara without further purification. Chemical materials such as hydrochloric acid (1 N HCl), calcium carbonate (CaCO₃), sodium metabisulphite (Na₂S₂O₅), and sodium hydroxide (NaOH) were purchased from CV Rudang, Medan, Indonesia.

2.2. Extraction of durian seed’s starch

Prior soaking for 12 h in CaCO₃, the skin of durian seed was removed and the seed then was cut into pieces. The pieces then were washed, pulverized, and squeezed. In order to optimize that all remaining starch was fully extracted, the residue was re-washed for 2-3 times or until the filtrate was clear. After 24 h precipitated, the filtrate then underwent decantation. The residue of decantation then was added 0.5% (w/v) aqueous solution of Na₂S₂O₅ for starch cleaning. The resulted wet starch then was dried in the convection oven (50°C for 48 h). The pure dried starch was ground and sieved with an 80-mesh size and stored in the plastic bag at room temperature for further use.

2.3. Production of dextrin and adhesive

Dextrinization was started with making 30% (w/v) suspension from 30 g starch and 100 ml distilled water. The HCl was titrated into starch suspension until the pH was 1. Liquefaction was carried out on 95°C for 35 minutes and then neutralized using 20% (w/w) NaOH. The resulted wet dextrin then was dried in the convection oven (50°C for 48 h). The dried dextrin was ground and sieved with an 80-mesh size and stored in the glass bottle at room temperature for further use.

For making adhesive, each dextrin should be converted into suspension first with ratio dextrin and water as solvent was set at 1:20 (w/v) for enable spraying using a spray gun. The suspension was stirred using magnetic stirrer and then heated in boiling water bath for 15 minutes to be homogenous. The resulted adhesive was stored at ambient temperature for further use.
2.4. Determination of basic properties of the dextrin’s adhesive

Specific gravity ($SG$) was measured using gravimetric method using picnometer and analytical balance. An empty and oven-dry picnometer was weighed ($W_1$), distilled water-filled picnometer was weight ($W_2$), and dextrin’s adhesive-filled picnometer was also weighed ($W_3$). The specific gravity was determined as in Equation (1)

$$SG = \frac{(W_3 - W_1)}{(W_2 - W_1)}$$  \hspace{1cm} (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 ($W_1$) was dried in the oven at $(103 \pm 2)^{\circ}C$ for 24 h until constant weight ($W_2$). The solid content was determined as in Equation (2)

$$SC = \frac{(W_2)}{(W_1)} \times 100$$  \hspace{1cm} (2)

Viscosity was measured using a viscometer with appropriate spindle. Sample of dextrin’s adhesive was placed in a 100 ml beaker glass, and measurement was conducted in room temperature ($25^{\circ}C$) using spindle with velocity in rpm (rotary per minute).

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 dextrin’s 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 UF resin and NaOH for PF in order to make curing. In this experiment, dextrin does not require specific condition like acidic or alkaline. Therefore simple method for determining gel time was applied. About 10 grams dextrin’s 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.

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

Clarity of the adhesive was performed by visual observation either using naked eye or microscope. Sample of the dextrin’s adhesive was poured onto a petri dish or a glass slide in order to form film layer. Observation of color and existence of alien objects such as granules or dust particles was carried out accurately using certain magnification. Further, detailed observation on size and dimension of dextrin’s particles were done with aid of photo-microscope (Zeiss) for measurement; color was analyzed using a chroma meter with parameters observation including $L$ (lightness), $a$ (redness), $b$ (yellowness). $L$ showed a value of color lightness while $a$ and $b$ was converted into value of chroma (saturation, vividness or dullness) and hue (how most of us perceive and a name of color according to rainbow color), respectively. Calculation of the hue was determined as in Equation (3)

$$hue = a \tan \left( \frac{b}{a} \right) \times \frac{180}{\pi}$$  \hspace{1cm} (3)

2.5. Making particleboard using dextrin’s durian seed and testing of the quality

Particleboard made of oil-palm trunk particles with dextrin’s durian seed as the binder was manufactured in laboratory. Target density was 0.75 g/cm$^3$, board dimension was 25 cm x 25 cm x 1 cm, and adhesive content was 10% based on oven-dried particle. Hot-press was set at 25 kgf/cm$^2$ and $150^{\circ}C$ for 30 minutes with 3 steps, namely first-press (10 minutes); breathing-press for releasing moisture and preventing blowing (10 minutes); and curing-press (10 minutes).
Bonding feasibility testing was applied by internal bonding evaluation using Japanese Industrial Standard (JIS) A 5908 (2003) using a Tensilon Universal Testing Machine (UTM). Calculation of internal bonding ($IB$) strength was determined by value maximum load ($P_{max}$) divided by size dimension of sample ($A$) as in Equation (4)

$$IB = \frac{P_{max}}{A}$$  

(4)

### 3. Results and discussions

Yield of starch made from durian seed was 7.53%. This value was lower compare to previous work which resulted around 9-10% [11] because of raw material itself and processing. Durian seed was varied depend on the species and origin. In addition durian seeds were collected as the residue of direct human consumption from durian fruit’s stores. Therefore, it was difficult to determine specific kind of the durian. Further, processing or extracting starch from durian seed involved sodium metabisulphite ($Na_2S_2O_5$) as cleaning agent. This chemical gave influence in the yield depend on the concentration used and soaking time. As the concentration increased the yield decreased while the longer soaking time the yield increased. In this experiment concentration of the cleaning agent was higher around 0.2-0.5% therefore the yield was lower. The concentration used of cleaning agent in this experiment was adopted from [13].

All the dextrins have $SG$ around 1 with dextrin of durian has the highest value (more than 1) as shown in Fig.1. It means all the dextrins tend to sink. The SC was in line with values of $SG$ as shown in Fig. 2. Values of SC were very low compare to synthetic resins, like formaldehyde based resins usually have 50-60% solid and isocyanate has very high SC up to 98-99%. The lower SC indeed, because dextrin should be dispersed in water, heated, and added some chemical additives to modify the properties [14]. Therefore when dextrin was used as the binder in wood adhesive system, it recommended increasing the adhesive content.

![Figure 1. SG of all the dextrins in this experiment](image1.png)

![Figure 2. SC of all the dextrins in this experiment](image2.png)

The gel time of durian seed dextrin showed the lowest compare to sago and cassava dextrin as shown in Fig. 3. By contrast, pH of durian seed dextrin exhibited the highest (tend to neutral) compare to sago and cassava dextrin because of termination of liquefaction stage using neutral condition.
Types of dextrin

| Types of dextrin | Form    | Color              | Dust particle |
|------------------|---------|--------------------|---------------|
| Durian seed      | Liquid  | White purple       | +             |
| Sago             | Liquid  | Brown              | +             |
| Cassava          | Liquid  | White brown        | +             |

Detailed observation using a photo-microscope and viscosity measurement using a viscometer resulted in granule shape and size/ dimension as well as viscosity as presented in Table 2.

Table 2. Further observation using photo-microscope of the dextrins in this experiment

| Types of dextrin | Granule shape                | Size diameter (µm) | Viscosity (cP) |
|------------------|------------------------------|--------------------|----------------|
| Durian seed      | Spherical                    | 1.09 ± 0.15        | 3.04           |
| Sago             | Oval                         | 1.06 ± 0.14        | 7.12           |
| Cassava          | Quite round up to spherical  | 1.36 ± 0.17        | 19.72          |

Color analysis using a chroma meter was carried out in both starch and dextrin. Comparison data were presented in Fig. 5 for starch and Fig.6 for dextrin.
Even though value of lightness (L) decreased after starches were converted into dextrin, the L of all starches and dextrins were classified as clear because both values closed to 100. For hue or our perception according to rainbow color, all the starches were grouped into yellow red (YR) because the hue values in the ranged 54-90. However after converted into dextrin, only durian seed dextrin was altered. It was classified into red (R) because the hue value became 50.3.

Unfortunately, feasibility testing of internal bonding (IB) generated various data. JIS standard required 3.06 kgf/cm$^2$ for IB strength. In this study, dextrin made from durian seed resulted in 0.74 ± 0.92 kgf/cm$^2$ although there was individual data showed up to 3.30 kgf/cm$^2$. Uneven distribution of adhesive presumably making less strength of IB.

### 4. Conclusions

For sum up, dextrin made from durian seed has been successfully synthesized. Even though there were some differences on basic characteristics of this dextrin with sago and cassava particularly color, for feasibility bonding dextrin made from durian seed seemed promising in point of view the potency and the feasibility.

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