Drying of Merbau (*Intsia bijuga*) Sawdust Extract: Effect of Temperature on the Quality of Natural Dye Product

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Abstract: *The textile sector leaves 80% of effluent untreated. Carcinogenic, mutagenic, and poisonous synthetic colors in textile waste streams harm the ecosystem. Biodegradable natural dyes are safer than synthetic dyes. Merbau sawdust is abundant in Papua but underutilized. Merbau wood (*Intsia bijuga*) has considerable tannin and dye potential. Tannin diffuses to the surface, coloring it brown in humid air or water. Merbau extract can be dried to make powdered natural dye. This study examines Merbau sawdust extract drying and natural dye quality at different temperatures. The drying process was carried out in a convective oven at 60, 75, and 90 °C. The data showed that the solid product of natural dye was already dried at 60–90 °C within 100 min. The content of tannin in the dried natural dye powder was analyzed by titration method. The tannin content in the solid product was 0.5–0.9 g tannin/g solid. The drying rate during the constant drying rate period is around 0.00137 to 0.00256 g/cm^2 min. The effect of drying temperature (60 to 90 °C) on the tannin degradation is insignificant, based on the titration method and FTIR analysis.*

Keywords: *drying; natural dye;* Intsia bijuga*; mass and heat transfer; tannin*

■ INTRODUCTION

The textile industry is one of the largest water-consuming industries and the most significant contributor to pollution in water bodies. Synthetic dyes are more widely used in the textile industry because they produce consistent colors for each batch production and do not easily fade when applied to the fabrics [1]. However, the chemicals contained in synthetic dyes can be harmful to the environment [2]. In the textile industry, untreated dye-containing wastewater potentially contributes up to 80% of all emissions [1-2]. In most of the waste generated from the textile industry, there are very high levels of biochemical oxygen demand (BOD) and chemical oxygen demand (COD). In addition, synthetic dyes can inhibit plant growth and photosynthesis and increase bioaccumulation, toxicity, mutagenic, and carcinogenic properties [3]. An alternative dye that is safe for the environment, non-toxic, and easily degraded in nature is needed.

Dye is a pigmented compound that colors a material or object [4-5]. Dyes are divided into two types, namely natural and synthetic dyes. Natural dyes are extracted from plants, animals, or microorganisms available in nature [5-6]. Meanwhile, synthetic dyes are made from artificial materials resulting from derivatives of hydrocarbon compounds such as benzene and naphthalene. The advantage of natural dyes is that they do not contain harmful chemicals and are biodegradable or decomposed naturally by the environment, so they do not harm the ecosystem. Natural dyes fade faster than synthetic dyes, so fixative compounds such as salt or vinegar are needed [7]. In addition, there is no specific natural dye production method to produce the same color composition each time it is made.

In contrast, the production method of synthetic dye can be adjusted to the desired color composition. However, the drawback is that synthetic dyes can produce carcinogenic, toxic, and mutagenic wastes, harming the living things around them. Therefore, it is necessary to empower natural dyes more [8].

Natural dyes are often obtained from plants with color pigments in their roots, stems, leaves, fruit, and skin [8-11]. There are two various ways to isolate the natural dyes, which are: (a) extraction of natural dyes with water or organic solvent at high or low temperatures for a specific time [12], and (b) extraction of natural dyes by hydrolyzing the material with acids or bases, according to its polarity.

Natural dyes can be used in various industries, such as food, textile, cosmetics, and pharmaceuticals [12-14]. Some examples of natural dyes that are often used in the textile industry are indigo (*Indigofera* sp.), soga bark (*Ceriops candolleana* Arn.), tegeran wood (*Cudraina javenensis*), and kesumba (*Bixa orellana*). To prevent the color on the textile from fading quickly, adding a dye fixator or mordant in the fabric dyeing process is necessary. Mordant can be selected from non-toxic materials to prevent environmental pollution [15]. Some of the most commonly used fixators are alum, borax, lime water, vinegar, and palm sugar.

Various studies have been conducted on numerous plants to produce natural dyes [8,10]. Many plants in Indonesia can be utilized as natural sources of dyes, such as Indigofera, turmeric, tegeran wood. Fatubun et al. [14] used eight different types of plants to create a diverse range of colors, including red, blue, yellow, and brown. Their innovative approach involves harnessing the natural pigments present in these plants to produce vibrant and varied hues. This method showcases the rich biodiversity of plant-based colorants and highlights the potential for sustainable and eco-friendly alternatives in various industries.

Moreover, Fauziyah and Hakim [15] utilized ten different types of plants to create a spectrum of colors, ranging from deep crimson to bright green and even shades like olive and lime. Their research highlights the extensive palette that natural sources can offer, showcasing variations from deep scarlet to vibrant emerald, demonstrating the diverse applications of plant-based coloration in various fields. In addition, The availability of multiple plants in Indonesia can be a potential for Indonesia to compete in the international market. One of the plants are widely available in Indonesia that can produce a natural brown color is the wood of the Merbau (*Intsia bijuga*).

Merbau trees commonly can grow to a height of more than 30 m. The wood of this tree is often used as a building material and as a wooden floor for ships. The Merbau tree, known as ironwood by the people of Papua, can produce multifunctional Merbau wood. This wood does not rot quickly if not in contact with soil and is stronger than teak [16]. Merbau wood has high economic value and has been used locally and exported. The Merbau tree is a type of tree commonly found both in Papua and Java [17]. Many timber mills and plywood industries in Papua utilize Merbau wood as raw material and produce sawdust as waste.

The speciality of Merbau timber is its very high tannin content. Tannins are a type of polyphenol that is found in many plant species, including trees, and serve to protect the plant from herbivores and other environmental stressors. Merbau wood, which comes from the *Intsia* spp. tree, contains a group of tannins known as proanthocyanidins [18]. These tannins are also commonly referred to as condensed tannins or polyflavonoid tannins. Merbau tree wood contains tannin pigment, which is brown. The pigments in these plants can be used as natural dyes with high economic value. Therefore, the tannins in Merbau sawdust can be extracted as a natural dye.

Tannins are polyphenols consisting of several hydroxyl groups which can be extracted from plants [19]. The extraction process is generally carried out using the Soxhlet or extractor. After the natural dyes are extracted from Merbau wood, the extracts can be dried to form natural dye powder using a dryer. The tannins in natural dyes will be easily damaged if exposed to high temperatures. Drying the extract to produce powder requires a relatively high temperature and quite a long time so it will affect the quality of the natural dyes produced. From this research, parameters of drying rate and drying conditions can relatively maintain the quality of the natural dye product. An analysis of the degradation of tannins during drying and the tannin content in natural dye powders is carried out to determine the temperature effect on the natural dye product quality.

■ EXPERIMENTAL SECTION

Materials

The materials used in this study are Merbau sawdust extract obtained from Gama Indigo, Yogyakarta, and Merbau sawdust obtained from CV Karui, Jayapura, Papua. Distilled water, $KMnO_4$ (99% purity Merck, Germany), indigo carmine (C.I.73015 Merck, Germany), and H_2SO_4 (95–97% purity Merck, Germany).

Instrumentation

The experimental rig used in this research consists of an oven equipped with thermometers to measure wet bulb and dry bulb temperatures. This oven has a temperature controller that can be adjusted according to the experimental requirements. Additionally, an exhaust system regulates air circulation within the oven, maintaining a stable experimental environment and ensuring consistent results. The oven unit is the Memmert UN 30 Universal model and it is equipped with a 32-L capacity and allows for temperature adjustments of up to 300°C. The oven is a key component in the research, allowing for precise control and monitoring of temperature conditions, thus enabling the collection of accurate and reliable data for further analysis in the study. FTIR unit is Shimadzu FTIR IR-Spirit Serial No A224158. The experimental rig is shown in Fig. 1.

Procedure

Drying

Washed and dried petri dishes were weighed on a digital analytical balance, and their mass was recorded. Each petri dish was filled with 10 mL Merbau extract using a volume pipette, weighed, and its initial mass sample was recorded.

The oven was turned on, and the dry bulb temperature was set to 60 °C. When the dry bulb temperature was stable, the four petri dishes containing the extracts were put into the oven and positioned, as shown in Fig. 1. The four samples were weighed at the time interval of 5 min until the sample mass was constant.

The petri dishes containing dried samples were removed from the oven and then cooled in a desiccator for 10 min. Petri dishes were weighed, and the results were recorded. The petri dishes were covered with aluminum foil and left at room temperature for 24 h. The petri dishes were dried again at 110 °C for 1 h and weighted as a dry weight. The above steps were repeated for temperatures of 75 and 90 °C. The experimental data was a mass of samples at various times for 60, 75, and 90 °C drying temperatures.

Tannin content analysis

The analyses of tannin content in plant extracts were

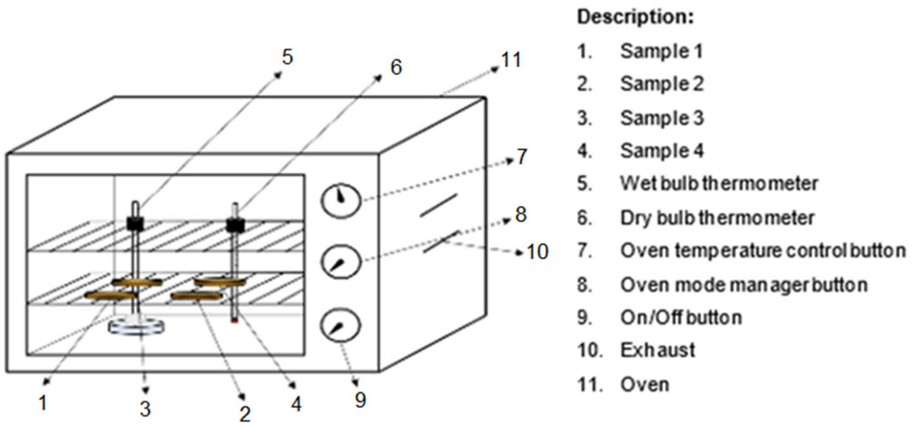

Fig 1. Experimental apparatus for drying

performed according to the International Pharmacopoeia and AOAC method in Kumari and Jain [20]. The preparation of 0.1 M $KMnO_4$ solution was mixing 0.79 g of $KMnO_4$ with 100 mL of distilled water in a beaker glass. The solution was heated for 15 min and left overnight at room temperature. The solution was filtered and added by distilled water up to 250 mL. Indigo carmine indicator solution was prepared by mixing 3 g of indigo carmine with 100 mL of distilled water and dissolving with 25 mL of concentrated H_2SO_4. The solution was heated for 15 min and left overnight at room temperature. The solution was then diluted until 500 mL with distilled water.

Tannin analysis was performed by titration. A blank solution (without extract sample) was prepared by adding 20 mL of the indigo carmine solution with 180 mL distilled water. The blank solution was titrated with $KMnO_4$ until the color changed from blue to yellow. Titration was carried out three times. A total of 10 mL of liquid Merbau sample was put into a 200 mL volumetric flask. As much as 20 mL of indigo carmine solution was put into a volumetric flask, and then distilled water was added until it reached 200 mL. The liquid sample solution with indigo carmine was titrated with $KMnO_4$ until the color changed from blue to reddish brown. Titration was carried out three times. A petri dish containing dried samples was added with sufficient distilled water to form a solution. The entire sample solution on the petri dish was put into a 200 mL volumetric flask. Then, 20 mL of indigo carmine solution and 180 mL of distilled water were added and shaken until homogeneous. A 50 mL sample solution with indigo carmine was titrated with 0.1 M $KMnO_4$ solution. The titration was carried out until the color changed from blue to reddish brown. The titration was repeated three times for each sample.

Tannin degradation

An empty petri dish was prepared, and the initial mass was recorded. Merbau extract as much as 10 mL was put into a petri dish. The mass of the petri dish containing Merbau sample was weighed. The petri dish containing the sample was put into the oven at a specific temperature to heat and dry for 20 min. After that, the petri dish containing the sample was removed, cooled, and then weighed. This step was repeated for heating and drying at 40, 60, 80, 100, and 120 min at the same position in the oven. The sample phase at each drying time was recorded.

The first sample (drying time 20 min) from the previous step was dissolved with sufficient distilled water in a petri dish. The tannin content in the solution was then analyzed using the described method. The experiment was conducted for heating and drying temperatures of 60, 75, and 90 °C.

FTIR analysis

A sample of dried tannin was also analyzed with its related functional groups (especially –OH and C–O) using a FTIR unit, that is Shimadzu FTIR IR-Spirit Serial No A224158. This type of FTIR unit does not need special sample preparation, in which a solid sample can be examined directly without further preparation. The dried tannin samples from several drying temperatures (60–100 °C) were examined.

Data analysis

The water content in the sample was calculated using Eq. (1a) for wet basis and Eq. (1b) for dry basis [21];

$$x=\frac{W_{st}-W_{sc}}{W_{s0}-W_0} \tag{1a}$$

$$x=\frac{W_{st}-W_{sc}}{W_{sc}-W_0} \tag{1b}$$

where x represents the water content in the sample (g H_2O/g wet sample), W_{st} represents the mass of the sample and container at time t min (g), W_{sc} represents the mass of the dry sample and container at the end of the drying process (g), W_{s0} is the mass of the initial sample and container (g), and W_0 is the mass of the empty container (g). The tannin content in the sample can be calculated using Eq. (2) [22];

$$\text{Tannin content}=\left(\frac{\text{g of tannin}}{\text{mL of sample}}\right)=\frac{(V_1-V_2)\times 0.004157\times FP}{V} \tag{2}$$

where FP denotes the dilution factor (200 mL/50 mL), V_1 denotes the volume of $KMnO_4$ used for sample titration (mL), V_2 denotes the volume of $KMnO_4$ used for blank titration (mL), and V denotes the volume of

Merbau sample dried on a petri dish (10 mL). One mL of 0.1 M $KMnO_4$ is equivalent to 0.004157 g of tannin.

Meanwhile, the drying rate N can be calculated using Eq. (3) [23].

$$N = -\frac{1}{A} \cdot \frac{dW_s}{dt} \tag{3}$$

In this case, W_s (g) represents the mass of the sample at any time (= $W_{st} - W_0$), A is the effective drying surface area (cm^2), and t is the drying time (min). During the constant drying rate, the curve W_s against drying time t is essentially a linear line, with its slope equal to drying rate N. The mass transfer coefficient (K_Y) in the constant drying rate period can be calculated by Eq. (4) and (5) [24];

$$N = K_Y (Y_s' - Y') \tag{4}$$

$$K_Y = \frac{N}{(Y_s' - Y')} \tag{5}$$

where N denotes the drying rate at a constant drying period (g H_2O/cm^2 min), K_Y denotes the gas phase mass transfer coefficient (g/cm^2 min ΔY), Y_s' is the saturated humidity of air at wet bulb temperature (g H_2O/g of dry air), and Y' is the humidity of the drying air (g H_2O/g of dry air). The Y_s' and Y' values can be calculated using Eq. (6-9) [24];

$$Y_s' = 0.622 \frac{P_{as}}{(P_t - P_{as})} \tag{6}$$

$$Y = \frac{\lambda_w Y_s' - 0.24(T_d - T_w)}{\lambda_w + 0.45(T_d - T_w)} \tag{7}$$

$$C_s = 0.24 + 0.45Y \tag{8}$$

$$Y' = Y_s' - \frac{C_s}{\lambda_w}(T_d - T_w) \tag{9}$$

where P_{as} denotes the saturated vapor pressure of water at wet bulb temperature (mmHg), P_t is the total system pressure (760 mmHg), C_s denotes humid heat, (Btu/lb °F), 0.24 and 0.45 are values of the specific heat of the air and water vapor (Btu/lb °F), respectively, λ_w denotes latent heat of vapor (Btu/lb), T_d denotes dry bulb temperature (°F), and T_w is the wet bulb temperature (°F). Eq. (10) and (11) estimate the heat transfer coefficient during constant drying rate (N) [24];

$$N = \frac{Q}{A \cdot \lambda_w} = \frac{hA(T_d - T_w)}{\lambda_w} \tag{10}$$

$$h = \frac{N \times \lambda_w}{(T_d - T_w)} \tag{11}$$

where N represents the constant drying rate (g H_2O/m^2 s), h is the convection heat transfer coefficient (W/m^2 °C), and Q denotes heat transfer rate (J/s).

The tannin degradation or content reduction rate is estimated by first-order reaction, as commonly used in degradation reactions (Eq. (12));

$$\frac{dM_{ta}}{dt} = -k_d M_{ta} \tag{12}$$

M_{ta} is the tannin content, and k_d is the degradation rate constant. Integration of Eq. (12) from t = 0 (initial condition with tannin content = M_{ta0}) results in Eq. (13) [25].

$$\ln\left(\frac{M_{ta}}{M_{ta0}}\right) = -k_d t \tag{13}$$

The degradation rate constant k_d can be expressed as the Arrhenius equation in this case.

RESULTS AND DISCUSSION

The variation of the drying process was the oven temperatures of 60, 75, and 90 °C. Fig. 2 shows the water content on a wet basis, and Fig. 3 shows the water content on a dry basis as a function of drying time. Fig. 2 presents a drastic decrease in water content in the initial period. Fig. 3 demonstrates that the dry basis water content linearly decreases with the time in the initial period of the drying process, known as a constant-rate period.

The drying process goes through three stages: the initial period, the constant-rate period, and the falling-rate period. The initial drying period occurs at several minutes. The constant drying rate follows after the initial drying period, where the sample mass decreases linearly with the drying time. The value of drying rate constant can be estimated from Fig. 2, which is based on Eq. (3). The value of the constant drying rate is used to calculate the mass transfer coefficient and the heat transfer coefficient.

From Fig. 4, it can be seen that the longer the drying time, the more the sample mass will decrease. The sample mass became constant after drying for 100 min at 60 °C, 60 min at 75 °C, and 40 min at 90 °C. Table 1 presents the value of the constant drying rate of 4 samples at 60, 75, and 90 °C. It shows that the sample

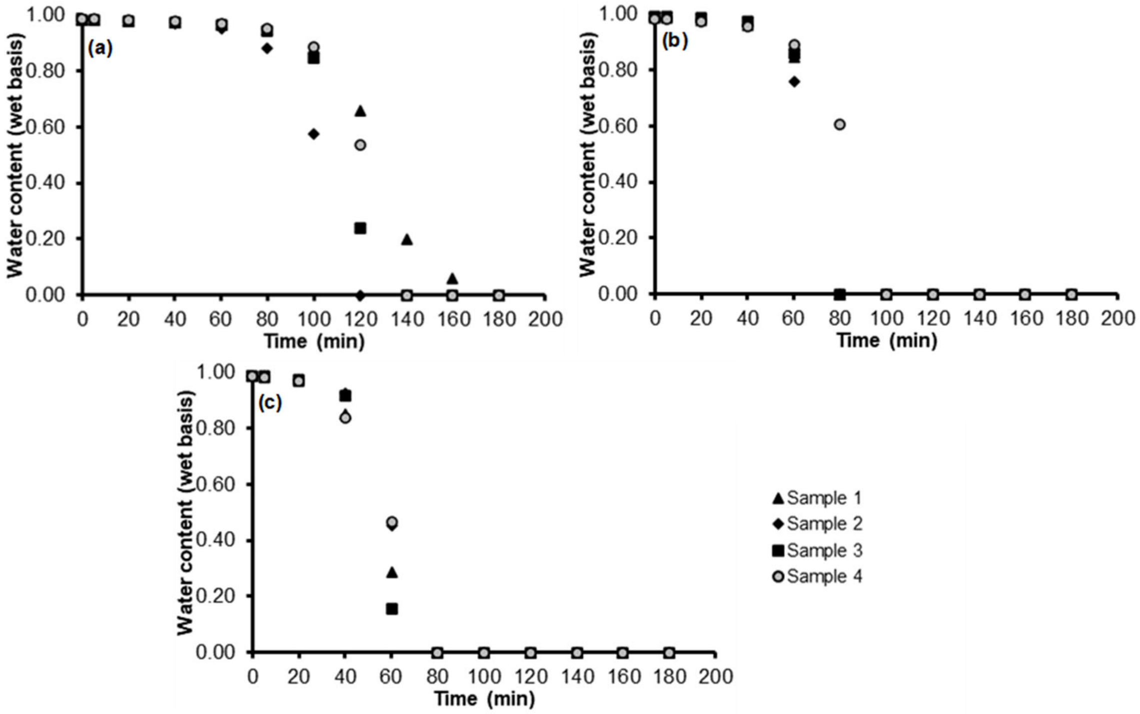

Fig 2. Water content (wet basis) at various times, temperature:(a) 60 °C, (b) 75 °C, and (c) 90 °C

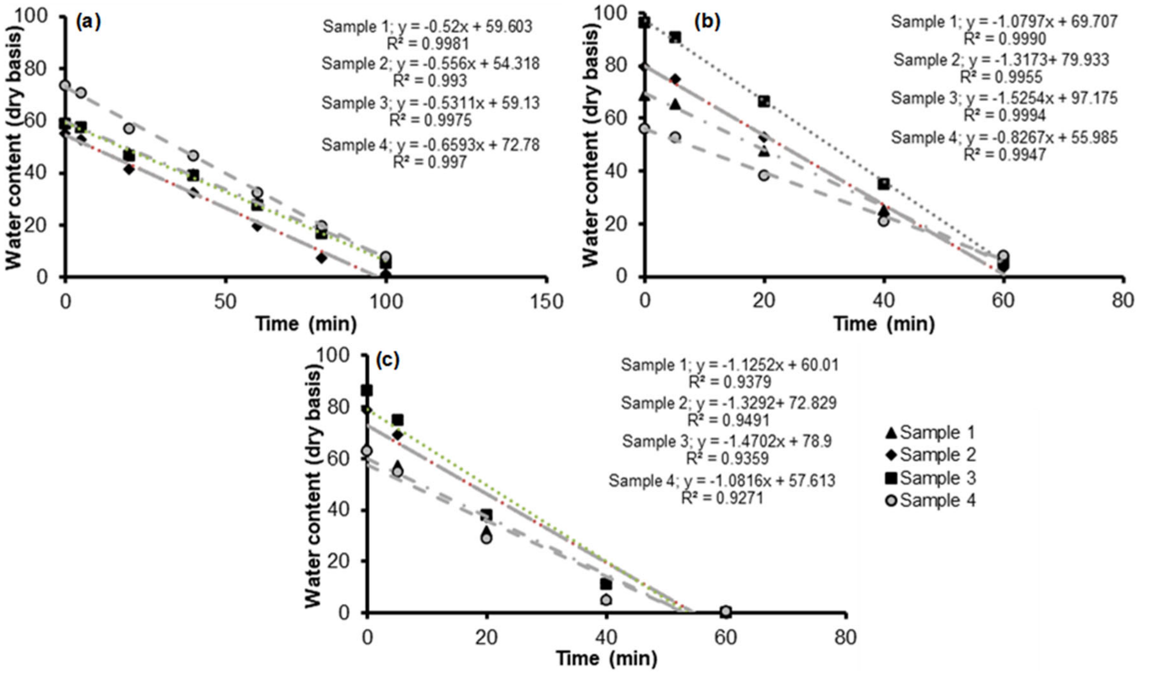

Fig 3. Water content (dry basis) as a time function, temperature: (a) 60 °C, (b) 75 °C, and (c) 90 °C

Table 1. Values of constant drying rate at 60 to 90 °C

Temperature (°C)	Sample 1	Sample 2	Sample 3	Sample 4	N avg (g/cm^2 min)
60	0.00131	0.00149	0.00134	0.00135	0.00137
75	0.00238	0.00249	0.00240	0.00221	0.00237
90	0.00265	0.00251	0.00254	0.00255	0.00256

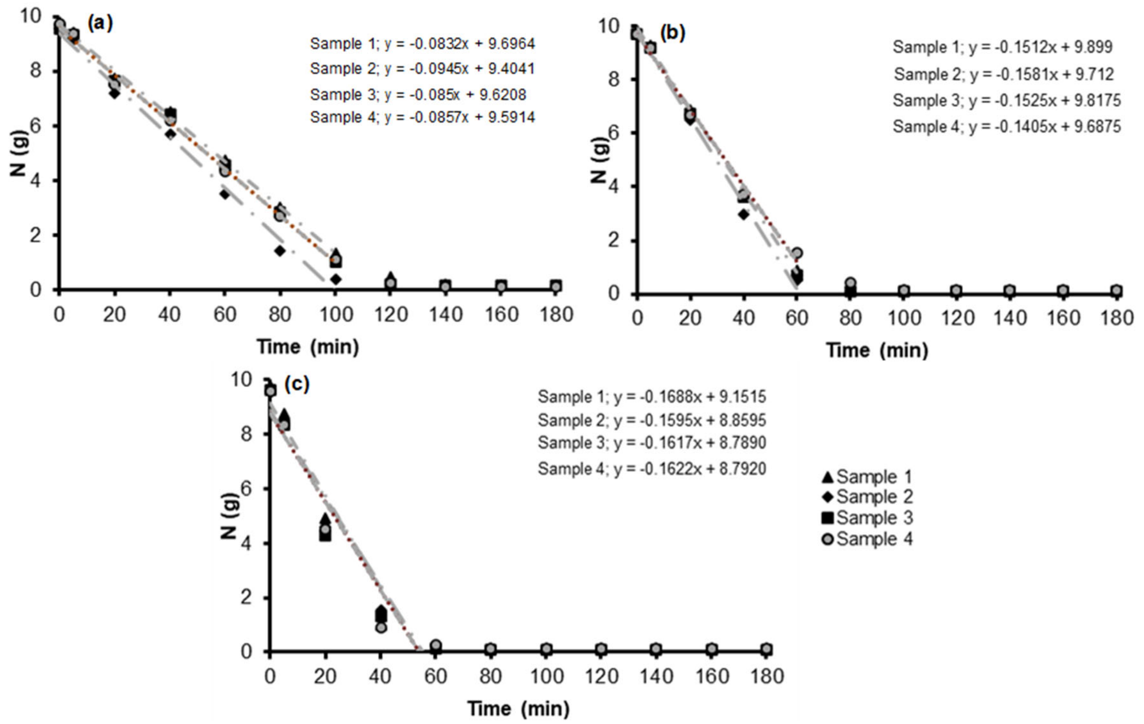

Fig 4. The sample mass as a time function at temperatures of (a) 60 °C, (b) 75 °C, and (c) 90 °C

position within the dryer tray does not affect the drying rate, so the value of the constant drying rate is taken as the average of 4 samples.

Based on Eq. (4-9), the average mass transfer coefficient at a constant drying rate and the average heat transfer coefficient can be calculated. Table 2 summarizes the values of mass transfer and heat transfer coefficients.

Ndukwu et al. [26] studied the drying of cocoyam slices using a drying oven, similar to this research. They found that the mass and heat transfer coefficients were around 1.01×10^{-6}–3.45×10^{-6} m/s and 1.18–3.58 W/m^{2}K, respectively. The heat transfer coefficient from this research is higher than that of Ndukwu et al [26]. For comparison, the K_Y unit in this research is converted into a m/s unit, which results in 1.43×10^{-5}–2.42×10^{-5} m/s. Table 3 compares heat and mass transfer coefficients between this research's and other research results. This phenomenon shows that the mass and heat transfer coefficients in this research are more or less within the range of other research results.

The tannin content in the four samples was analyzed by the volumetric method. Table 4 presents the values of tannin content for all drying temperatures.

Table 3. Comparison of mass and heat transfer coefficients

	K_Y (m/s)	$\bar{h}$ (W/m^2 °C)
This research	1.43×10^{-5}–2.42×10^{-5}	11.06–14.64
Ndukwu et al. [26]	1.01×10^{-6}–3.45×10^{-6}	1.18–3.58
Kumar et al. [27]	-	33.35–58.0
Chen et al. [28]	-	19.96

Table 2. Mass transfer coefficient and heat transfer coefficient at various temperatures

Drying temperature (°C)	$\overline{K_Y}$ (g H_2O/cm^2 min)	$\bar{h}$ (W/m^2 °C)
60	0.0859	14.64
75	0.1067	11.06
90	0.1452	12.29

Table 4. The tannin content in the dried sample (after the drying process)

Temperature (°C)	Sample 1	Sample 2	Sample 3	Sample 4	Average (g/g)
60	0.556	0.523	0.561	0.684	0.581
75	0.653	0.762	0.906	0.533	0.714
90	0.593	0.816	0.786	0.593	0.697

Based on the value of tannin content at each drying temperature, as shown in Table 4, the highest average tannin content was obtained at a drying temperature of 75 °C. However, the value of tannin content much varied, which was around 0.5 to 0.9 g tannin/g. Hence, the effect of drying temperature cannot be observed from merely tannin content at various temperatures. The effect of heating on the tannin quality or content over time was then investigated by heating and drying samples for 20 to 120 min at temperatures of 60, 75, and 90 °C. Fig. 5 presents tannin content (in 10 mL initial Merbau extract) at various times.

The tannin content in sample data can be used to evaluate the rate of tannin degradation based on Eq. (14). Three lines in Fig. 5 show the estimated correlation between tannin content and drying time at temperatures of 60, 75, and 90 °C. The degradation rate constant is given in Table 5, which is quite small. This phenomenon indicates that the tannin degradation rate is very low.

This rate constant k_d is expressed in the Arrhenius equation, Eq. (14):

$$k_d = 1.0061 \exp(-15.11/RT)\,\text{min}^{-1} \tag{14}$$

It shows that activation energy = 15.11 J/mol is small; therefore, the temperature effect on tannin degradation is insignificant.

The effect of temperature on the quality of tannin content was then investigated through the concentration or content of functional groups within the tannin sample drying at various temperatures. In this case, the corresponding functional groups in tannin are hydroxyl (O–H) and carbonyl (C=O and C–O) functional groups. Examination of these functional groups was carried out using FTIR.

Fig. 6 depicts the tannic acid spectrum, which exhibits high absorption between 3500 and 2900 cm^{-1}, with a broad band centered at 3262 cm^{-1}. The hydroxyl groups (O–H) H-bonded broadly and powerfully, and the C–H are responsible for this band (aromatic medium). A strong peak caused by the alkane medium (C–H) is seen at 2718 cm^{-1}. The sharp and robust peak between 2920 and 2807 cm^{-1} corresponds to the symmetric and

Table 5. The tannin degradation rate constant

Temperature (°C)	Constant k_d (min^{-1})
60	0.000673
75	0.000712
90	0.001130

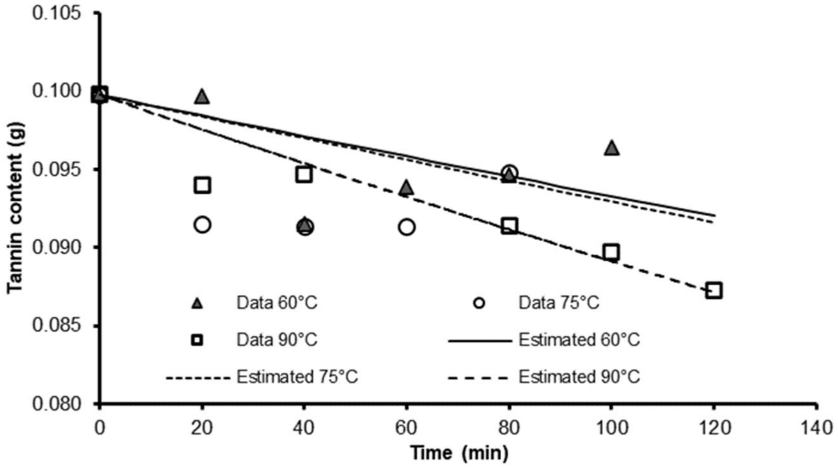

Fig 5. The tannin content in 10 mL sample at various times and temperatures

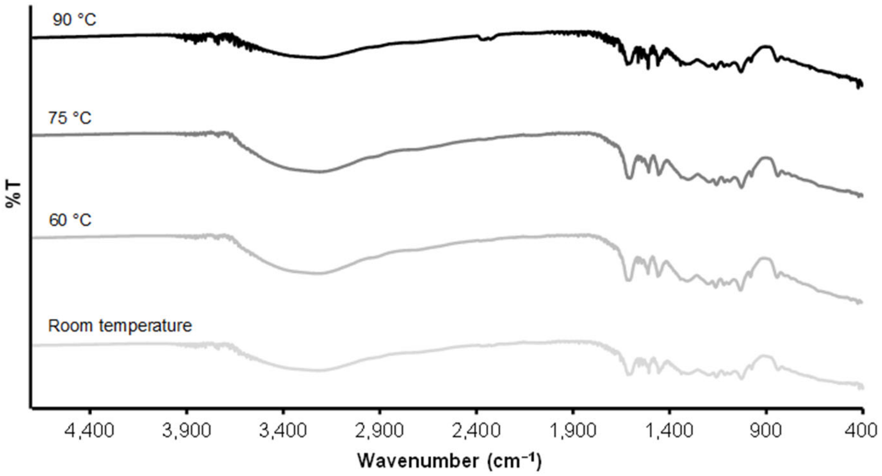

Fig 6. FTIR analysis results of samples at various drying temperatures

antisymmetric C–H stretching vibrations of the CH_2 and CH_3 groups, respectively. Due to the signal properties of carbonyl groups C=O stretching (1610 cm^{-1}) and C–O, tannic acid contains certain aromatic esters (1160 cm^{-1}).

Further experiments were conducted to investigate the thermal degradation of tannin. Some tannin solution was dried at several temperatures, i.e., from 60 to 100 °C, until solid tannin was produced. The drying process was around 3 h. FTIR then examined this solid tannin product. Based on the FTIR results of absorbance values for functional groups, the concentration of O–H at 3200 cm^{-1}, C–O at 1160 cm^{-1}, and aromatic C–H at 1450 cm^{-1} were estimated by their absorbance values. In this case, the amount of aromatic C–H was assumed to be constant along the drying process, irrespective of the drying temperature.

On the other hand, hydroxyls in tannin probably occurred to react to give the C–O functional group, as follows slowly, Eq. (15).

$$-C-OH+HO-C-\rightarrow -C-O-C-+H_2O \qquad (15)$$

Fig. 7 depicts the O–H and C–O absorbance ratio, compared to aromatic C–H absorbance, based on FTIR results at 60 to 100 °C. Fig. 6 indicates that during the drying process, there was a slight decrease in the O–H group when the drying temperature rose, and according to the reaction in Eq. (15), the C–O group also slowly but steadily increased. However, a small reduction of O–H and an increase in the C–O group have little effect on

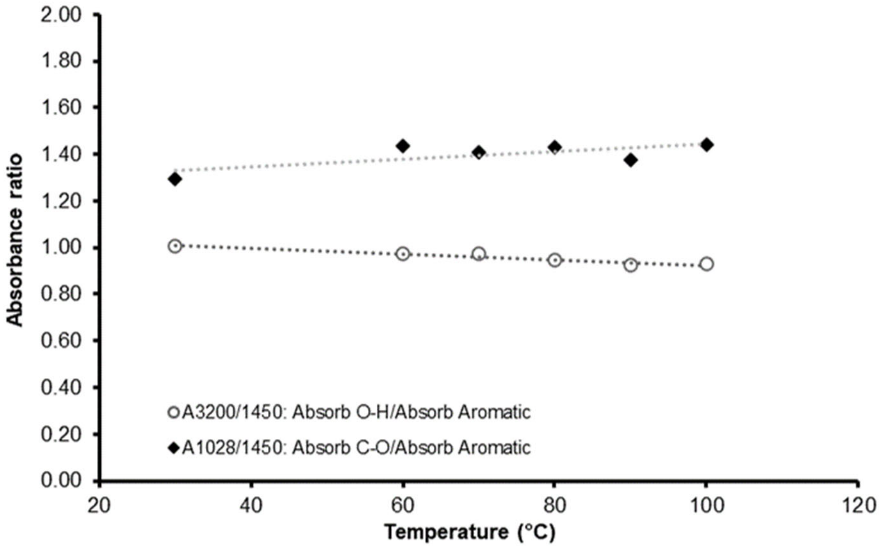

Fig 7. Absorbance changes of tannin hydroxyl and C–O functional groups at various drying temperatures

tannin quality. As shown in Table 4, the value of tannin content has significant variation, irrespective of drying temperature. It can be concluded that drying temperature in the range of 60 to 90 °C has little effect on the quality of natural dye products.

CONCLUSION

In this study, several conclusions can be drawn. During the constant drying period, the drying rate is around 0.00137 to 0.00256 g/cm^2 min, in which the tannin solid product is already dry within 100 min at 60–90 °C. The tannin content in the dried product is around 0.5 to 0.9 g tannin/g solid. The drying temperature has little effect on the quality of tannin products. Observation of the absorbance intensity of hydroxyl and carbonyl functional groups using FTIR indicates that higher drying temperature tends to reduce the amount of hydroxyl group slightly and to increase the number of carbonyl groups. At drying temperatures of 60 to 90 °C, the average mass transfer coefficient, K_Y, is 0.0859–0.1452 g H_2O/cm^2 min and the average heat transfer coefficient, h, is 11.06–14.64 W/m^2 °C. Both titration method and FTIR analysis show that the effect of drying temperature (60 to 90 °C) on tannin degradation is insignificant.

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AUTHOR CONTRIBUTIONS

Aswati Mindaryani, Sandrina Christine Michelin Silalahi, and Vincent Sutresno Hadi Sujoto conducted the experiment, Aswati Mindaryani, Vincent Sutresno Hadi Sujoto, Himawan Tri Bayu Murti Petrus wrote and revised the manuscript, Edia Rahayuningsih revised the manuscript. All authors agreed to the final version of this manuscript.

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