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Research article

The enhancement of sappanwood extract drying with foaming agent

under different temperature

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Abstract: Sappanwood (*Caesalpinia sappan* Linn) contains brazilin, a natural antioxidant. It can be extracted and dried to obtain a dry extract powder. However, sappanwood extract drying is difficult due to its tendency to form a jelly-like structure, which strongly traps water molecules. This research studies the effect of foaming agents (egg albumin and gum Arabic) as well as the drying temperatures (40, 60, and 80 °C) on the drying kinetics and physicochemical properties of the sappanwood extract powder. The water removal can be well expressed by the Page model. The addition of a foaming agent as well as the increase in drying temperature significantly speed up the drying process. However, at a temperature of 80°C, the effect of the foaming agents was less significant, and the dry extract became dark brown due to the occurrence of the Maillard reaction. By considering those trade-off phenomena, optimization with response surface methodology (RSM) was performed. The results indicated that sappanwood extract could be fully dried using a mixture of 5% egg albumin and 25% gum Arabic as the foaming agent at 64.1 °C, the in just 64.7 minutes or 7 times shorter compared to the drying without foaming agent. Under these drying conditions, the total phenolic compound retention was up to 87.25%.

1. Introduction

Sappanwood (*Caesalpinia sappan* Linn) is a terrestrial plant that belongs to the Leguminosae family and originates from Southeast Asia, China, and Japan. Numerous important functions of this wood plant have led to its cultivation in wider parts of the world, such as Africa, Europe, as well as North America and Latin America regions. Generally, the wood part is not only used in the preparation of traditional foods and beverages, but its superior health benefits have also driven its various applications in traditional medicines [1]. Sappanwood is also well-known as a good source of secondary metabolites, namely, brazilin and sappanchalcone, which both exhibit strong antioxidant activity [2]. Besides, the sappanwood extract also possesses antimicrobial activity that enables its utilization as a food preservative [1], which was specifically introduced into cooked sausages [3]. The reddish color of brazilein not only makes it an excellent natural food colorant such as a colorant for fish paste [4] and salted eggs [5], but it shows its potential application as a natural dye for the sensitized solar cells [6]. Furthermore, sappanwood extract has been reported to demonstrate the antidiabetic activity by controlling blood glucose levels [7].

As the major secondary metabolite of sappanwood, brazilin, which is a colorless phenolic compound, is usually extracted from sappan heartwood shavings using highly-polar solvents such as water, methanol, ethanol, or a mixture of methanol and dichloromethane [1]. Unfortunately, brazilin was reported to be very sensitive to the oxidation process due to direct exposure to air, heat, light, and pH [8]. Oxidation of the hydroxyl groups in its structure causes these to turn into carboxyl groups, causing the structural transformation and resulting in a new colored compound, namely brazilein. To extend the shelf-life and storage time as well as to maintain the valuable components, sappanwood extract needs to be converted into a more stable form such as dry powder. However, the water extract of sappan heartwood shavings is difficult to be dried due to its high hydrophilicity. The extract can easily imbibe water and form an extremely sticky jelly-like product. To overcome this problem, a foaming agent can be added to the extract that functions to break the linkage between the water molecules and sappanwood extract. As a result, the drying process is faster and results in a more stable dry powder with a longer storage period and higher secondary metabolite retention [9,10].

Foam mat drying is a process that is used to convert a liquid or semi-solid compound to a stable foam by the addition of a foaming and/or stabilizing agent. The foams, which are dispersions of gas in solid or liquid, are stabilized using a surface-active agent or surfactant [11]. The foamy liquid is then dried following the thin-layer principle using a hot air dryer and further ground to form a powder. Therefore, foam mat drying has been regarded as an easy and economical drying process [11]. The drying can be performed at lower temperatures and, consequently, appropriate for the dehydration of heat-sensitive products [9,10]. Foam mat drying resulted in the stable foam that enhanced the surface area of drying. While the larger surface area makes the drying rate in foam mat drying faster than any other drying kinetics [12].

In the foam mat drying process, the foaming agent plays an important role to reduce the surface tension between the gas and liquid phase. In addition, the foaming agent also increases the surface area that is exposed to the drying air, which finally speeds up the drying rate. The currently available foaming agent is usually originated from the protein, gum, or emulsifiers group. The selection of a foaming agent and its formulation will affect both the drying kinetics and the characteristics of the products. Thus, foam mat drying has been applied to produce stable milk powder, carrageenan powder, yogurt, and apple, papaya, and roselle extracts [9–11]. To the best of our knowledge, the application of foam mat drying for improving sappanwood extract powder quality has not been investigated. This study aims to investigate the effect of foaming agent formulation (egg albumin and gum Arabic), and the drying temperatures on the drying kinetics and physicochemical properties of the sappanwood extract powder. The egg albumin and gum Arabic contain protein that can result in stable foam in the drying process [13]. However, the high protein content can lead to the Maillard reaction [14]. This study combined egg albumin and gum Arabic to find the fastest drying process with high-quality product.

2. Materials and methods

2.1. Materials

The sappan heartwood shavings $(1.5 \times 1.5 \text{ cm} \text{ and } 0.2 \text{ cm} \text{ thickness})$ were collected from a local Herbal Market in Karawang, Indonesia. Distilled water was used as the solvent for the sappanwood extraction. Food-grade egg albumin and gum Arabic (derived from *Acacia Senegal* L.) were used for the foam formulation. Analytical grade Folin–Ciocalteu reagent, Na₂CO₃, gallic acid standard compound, and ethanol (purity \geq 99% w/w) were purchased from Merck (Darmstadt, Germany) and were directly used for the total phenolic compound analysis.

2.2. Methods

Figure 1 illustrates the schematic diagram of the sappanwood extract manufacturing study, which comprises sequential unit operation steps, namely, sappanwood extraction, foam mat drying, moisture content observation, sappanwood extract characterization, drying kinetics analysis, and optimization of foam mat drying process. In principle, the study began with experimental work, followed by the drying kinetics model selection and validation.

2.3. Preparation of sappanwood extract

In this work, water was selected as the solvent for the extraction of phenolic compounds from the heartwood of the sappan tree [15]. One-hundred grams of sappanwood shavings was introduced into 100 mL of distilled water at 70 °C in a 500-mL glass beaker. The mixture was continuously stirred at 150 rpm for 2 h to allow the release of the phenolic compounds from the heartwood of the sappan tree. Next, the red-colored liquid containing the sappanwood crude extract was separated from the exhausted sappanwood using a Whattman filter paper (series number 1) with the assistance of a vacuum pump.

2.4. Foam mat drying

Egg albumin and gum Arabic as the foaming agents were mixed with the crude sappanwood extract. The composition of foaming agents was selected where the egg albumin in the formulation

cannot be upper 30%. Higher egg albumin led to viscous mixture that was difficult to be processed. For this study, the first composition is 20% (w/w) egg albumin and 10% (w/w) gum Arabic called as S2. The mixture was then whipped using a hand mixer (2500 rpm) for 15 min to form sappanwood extract foam, and the resulting foam was placed onto a thin aluminum tray (diameter, 10 cm; thickness, 0.2 cm). The drying process was carried out in an electric oven at three different temperatures (40, 60, and 80°C) by employing drying air at a constant superficial velocity of 2 m/s. The sample was weighed every 10 min for a total drying time of 50 min for the determination of the moisture reduction. In addition, the total phenolic content of the sample was also quantified. The procedure was repeated for different foaming agent compositions/formulations such as 30% (w/w) egg albumin (S3), 10% (w/w) egg albumin and 20% (w/w) gum Arabic (S4), and 30% (w/w) gum Arabic (S5). As a comparison, the sappanwood extract without foaming agents called S1 was also performed for this procedure. In doing so, the amount of water was also added based on water balance estimation so each formulation had the same moisture content 90% (wet basis).



Figure 1. The sappanwood extract manufacturing study.

2.5.1. Moisture ratio

Moisture content was observed every 10 min by gravimetric method for 50 min. The observation was conducted twice every sampling time. The data were converted into dimensionless moisture content (moisture ratio). The moisture ratio, MR, was calculated using the following equation:

$$MR = \frac{(X_t - X_e)}{(X_0 - X_e)} \tag{1}$$

where X_t is the moisture content at a given time (t), X_0 is initial the moisture content (at t = 0), and Xe is the equilibrium moisture content. The X_t , X_0 , and Xe are expressed on a wet basis. The equilibrium moisture content (Xe) was evaluated by drying of the sappanwood extract sample until the achievement of its constant weight.

2.5.2. Thin-layer model

Given that the foaming agent and sappanwood extract mixture was placed on a 0.2-cm thick aluminum bed, thin-layer drying models could be expected to adequately describe the moisture removal. The thin-layer drying models chosen in this work are tabulated in Eq. (2) to Eq. (5). [16].

|--|

Page model
$$MR = exp^{-kt^n}$$
 (3)

Handerson-Pabis model
$$MR = a exp^{-kt}$$
 (4)

Logarithmic model
$$MR = a \exp^{-kt} + c$$
 (5)

The drying rate constant (min–1) is represented by the k value, whereas n, a, and c are the drying kinetic model's constants (Eq. (2) to Eq. (5)). To obtain all the model's constants, nonlinear regression was performed using Polymath Educational 6.0. The coefficient of determination (\mathbb{R}^2) and the root-mean-square deviation (RMSD) evaluation was subjected to the chosen thin-layer drying models presented in Eq. (2) to Eq. (5) to obtain the best model. Then, the best drying kinetic model was used to estimate the drying time (from an initial moisture content of 90% wet basis or 9 g water/g dry solid to a final moisture content of 12% wet basis or 0.13 g water/g dry solid).

2.6. Analysis of dried sappanwood extract

2.6.1. Scanning electron microscopy analysis

Scanning electron microscopy (SEM) was performed on the sappanwood extract powder using an electron microscope (JEOL JSM-6510LA) to observe its microstructure. The dried sappanwood extracts were mounted on an aluminum disk and coated with gold in a sputtering apparatus. To enable a thorough image observation, the sappanwood extract powder was scanned at $5,000 \times$ magnification.

2.6.2. Color measurement

The changes in the color of the crude and the dried sappanwood extracts were observed using a Chroma Meter (CR-300, Minolta Co., Ltd., Osaka, Japan) by assessing the following parameters: L* (lightness), a* (redness and greenness), and b* (yellowness and blueness) [9]. Then, the total color characteristic difference between the fresh and dried sappanwood extracts (ΔE) was calculated according to Equation 6.

$$\Delta E = \sqrt{(\Delta L^*)^2 + (\Delta a^*)^2 + (\Delta b^*)^2}$$
(6)

2.6.3. Fourier-transform infrared spectroscopy (FTIR) analysis

The Fourier transform infrared (FTIR) of the dried sappanwood extracts were recorded using a Frontier spectrometer (PerkinElmer, USA). The wavenumber employed for the observation of the functional groups existing in the extract ranged from 4,000 to 400 cm⁻¹.

2.6.4. Total phenolic compounds

The total phenolic compounds in the samples of sappanwood extract powder were determined using the Folin–Ciocalteu colorimetric method every 10 min [17]. The sample was diluted and reacted with Folin–Ciocalteu reagent for 2 h at room temperature (27–29 °C). The absorbance of the solution was measured at 765 nm using a UV-Vis spectrophotometer (GENESYS[™] 150 UV-Vis). Then, the observed absorbance was used to calculate the total phenolic compounds (as mg of gallic acid equivalents) by referring to the standard curve (concentration vs. absorbance) of the gallic acid solution.

A different drying condition can result in a different degradation rate of a specific compound contained in a certain food material. The degradation of the total phenolic compounds of goji berry [18], yacon [19], and kiwi [20] are well described by the first-order reaction. By considering the wide applications of this degradation kinetics approach, the first-order reaction will also be applied to describe the degradation kinetics of the total phenolic compounds in the sappanwood extract. Equation (7) represents the first-order reaction:

$$A_t = A_0 e^{-kt} \tag{7}$$

where A_t is the concentration of total phenolic compounds at a given drying time (mg of gallic acid equivalent/g dry sappanwood extract), t is the drying time (min), and k is the degradation rate constant of total phenolic compounds (min⁻¹). To evaluate the accuracy of the model, the model was fitted with the experimental data based on the coefficient of determination (R²) value, for which an R² value closer to 1 is desired.

2.6.5. Optimization using response surface methodology (RSM)

The process and response variables were correlated and optimized using RSM. The drying temperature (A) and foam formulation (B) were selected as process variables. Meanwhile, the total phenolic compounds (Y) were chosen as the response variable. The optimization study used the central composite design (CCD) by employing 10 runs. The correlation between the factors and responses was

(8)

Statistical analysis and optimization were performed using Minitab® (Version 19-Trial version, Minitab, LLC, Pennsylvania, USA).

where, Y is the response, γ_0 is the intercept, γ_1 and γ_2 are coefficients of linear effect, γ_{11} and

 γ_{22} are coefficients of quadratic effect, and γ_{12} is a coefficient of double interaction.

 $Y = \gamma_0 + \gamma_1 A + \gamma_2 B + \gamma_{11} A^2 + \gamma_{22} B^2 + \gamma_{12} A B$

3. **Results and discussion**

3.1. Dried sappanwood extract microstructure

expressed as a second-order polynomial model [21]:

The microstructure of the dried sappanwood extracts is presented in Figure 2. The dry sappanwood extract without the addition of foaming agents (S1 sample) exhibits a smoother surface than the other extracts, see Figure 2 part A. On the other hand, the surface of the S2-S5 dried sappanwood extracts presented in Figures 2 parts B to E possesses a rougher surface, which may be attributed to the presence of numerous wide-thin flakes derived from the foaming agents.

During foam mat drying, a porous structure is developed from the foaming agents, which enlarges the surface area for easier moisture removal. As a consequence, the drying rate is significantly enhanced [9,11]. However, after a certain period, the pores in the thin structures were found to reduce due to the high tendency of the bubbles to coalesce with their neighboring bubbles, resulting in the formation of larger bubbles and the creation of more drainage. As seen in the S3 sample, the pores are deeper and the number of pores is more than those in the S2, S4, and S5 samples, despite the flake thickness is very similar. The small initial bubble size of the S3 sample increase the foam stability by longer foam drainage as illustrated in Figure 2 part C. The foam drainage could occur when the foam merges with other foams, resulting in a larger-thin foam structure that is easily broken [11]. In this study, the addition of egg albumin as the foaming agent in the S3 sample improved the foam stability due to the increase in lamellae interfacial viscoelasticity and form the small initial bubbles. This phenomenon also occurred in a previous study, the egg albumin exhibits smaller bubble sizes and higher interfacial elasticity than the whey protein [22]. Due to the continuous removal of water from the sappanwood extract during the foam mat drying process, the large polysaccharide structure of the gum Arabic formed a more viscous solution that hindered foam formation and, subsequently, restricted the formation of even pores on the dried extract surface within the range of the studied temperatures (S5).

3.2. Dried sappanwood extract color

The color measurement was carried out to study the effect of foam formulation and drying temperature on the color of the sappanwood extract. The color parameters, namely, lightness (L^*) , redness and greenness (a^{*}), yellowness and blueness (b^{*}), and total color difference (ΔE) are summarized in Table 1. The sappanwood extract before foam mat drying showed different L*, a*, and b* values. The sappanwood extract without the addition of foaming agents (S1 sample) showed the lowest L* value, indicating that the sample was the darkest at the beginning. A similar observation was reported by Gao et al., wherein the initial color of a sample was strongly dependent on the nature of the foaming agents used [23].





Color analysis revealed that all sappanwood extract samples suffered from the reduction of a^{*} and b^{*} values after the drying process, which indicated the decline of their redness and yellowness. As expected, the total color characteristics difference between the fresh and dried sappanwood extract (ΔE)

values increased with elevated drying temperatures. The highest ΔE value was observed for sappanwood extracts obtained from foam mat drying at 80 °C. This observation confirmed that at a higher temperature, nonenzymatic browning (Maillard reaction) between the amino group of the egg albumin and aldehyde of the gum Arabic was more pronounced. This reaction produced unstable intermediates, resulting in a group of brownish substances of different molecular compositions, which are called melanoidins [24]. Therefore, foam mat drying at 80 °C cannot be recommended due to the darkening of products. This finding suggested that the total color difference was the highest and clearly explained the reduction of color quality. Brazilin, as the natural pigment of sappanwood extract, is sensitive to air, heat, light, and pH [7]. The introduction of heat in the drying process leads to browning via nonenzymatic (caramelization or Maillard reaction). Caramelization is a nonenzymatic reaction that happens when carbohydrates or sugars in food are heated. This phenomenon resembles the process of water molecule removal from sugar molecules and is followed by isomerization and polymerization steps. The Maillard reaction can also take place at room temperature but at a slower rate [25]. Therefore, at a lower drying temperature (at 40 °C), the value of total color difference was the lowest, which ranged from 10 to 21. The higher browning process under higher temperatures was also found in natural pigments such as capsanthin in red bell pepper [26], anthocyanin in plum puree [27], and anthocyanin in roselle [9]. In addition, the color changes can also be the result of the natural pigment's degradation during thermal processing [27].

Based on Table 1, the different foam formulations (S1 to S5) resulted indifferent values of ΔE . The presence of egg albumin could promote the thermally induced browning process. However, the addition of gum Arabic slightly inhibits the browning phenomenon (Maillard reaction). For example, at S3 with 30% egg albumin only, which is a protein foaming agent, a higher ΔE value was obtained, ranging from 17 to 35. This result agrees with the previous report on the color (anthocyanins) degradation of roselle extracts [9]. As expected, the color degradation rate constant of the samples with the addition of foaming agents was 1.4–2.5 times higher than that without foaming agents. Indeed, the Maillard reaction depends on factors such as temperature, pH, time, nature of the reactant (type of sugar and protein), and the reactant concentration [28]. The addition of protein-rich foaming agents significantly enhanced the rate of the Maillard reaction.

3.3. FTIR profile of the sappanwood extracts

The FTIR analysis was carried out to investigate possible interactions between the sappanwood extract and the foaming agents by comparing the FTIR spectra of the sappan heartwood extract with the addition of foaming agents (S2–S5 samples) with the FTIR spectrum of the sappan heartwood extract without the addition of foaming agents (S1 sample), see Figure 3. Figure 3 part A exhibits the FTIR spectrum of the control sample (S1). Meanwhile, Figures 3 parts B to E present the spectra of the sappanwood extract samples with the addition of foaming agents, which are S2–S5, respectively. Furthermore, Figures 3 parts A to E also demonstrate an obvious transmittance reduction of the OH vibration at around 3300–3400 cm⁻¹ of the sappanwood extracts obtained from foam mat drying at 40°C–80 °C for 120 min. The largest transmittance reduction of the OH vibration was observed for sappanwood extracts dried at the highest temperatures (80 °C). The OH group transmittance reduction indicated a decline in the number of OH groups in the sappanwood extracts, which could be associated with the reduction of the phenolic compound due to direct heat exposure during the drying process [29].

	Before dry	ing	Drying at 40	°C	Drying at 60	°C	Drying at 80	°C
61								
51	L*:	21	L*:	27	L*:	27	L*:	30
	a*:	39	a*:	34	a*:	24	a*:	27
	b*:	26	b*:	20	b*:	12	b*:	3
			ΔE :	10	ΔE :	21	ΔE :	27
52	L*:	29	L*:	24	L*:	17	L*:	15
	a*:	42	a*:	22	a*:	21	a*:	20
	b*:	10	b*:	11	b*:	6	b*:	15
			ΔE :	21	ΔE :	25	ΔE :	27
S3								
	L*:	33	L*:	23	L*:	22	L*:	16
	a*:	45	a*:	32	a*:	18	a*:	17
	b*:	19	b*:	15	b*:	4	b*:	7
			ΔE :	17	ΔE :	33	ΔE :	35
S4			\bigcirc					
	L*:	38	L*:	36	L*:	35	L*:	33
	a*:	39	a*:	27	a*:	24	a*:	14
	b*:	6	b*:	3	b*:	0	b*:	0
		38	ΔE :	13	ΔE :	16	ΔE :	26
S5								
	L*:	38	L*:	44	L*:	43	L*:	50
	a*:	39	a*:	20	a*:	18	a*:	19
	b*:	10	b*:	8	b*:	1	b*:	7
			ΔE :	20	ΔE :	23	ΔE :	24

Table 1. Appearance of sappanwood extract by different foam formulations (S1,	S2,	S3,
S4, and S5) and drying temperatures (40, 60, and 80 °C).		

As seen in Figures 3 parts B to E, the Amide III vibration (C-N stretching, N-H bending) appears at a wavenumber range of 1,200 cm⁻¹. The existence of Amide III vibration in the spectra of samples S2–S5 confirms the presence of protein in the egg albumin and gum Arabic, which was used as a foaming agent during the preparation of both extracts. No Amide III vibration appeared in the sappanwood extract without the addition of foaming agents (Figure 3 part A). As expected, a higher drying temperature caused the transmittance of the Amide III vibration to decline. Furthermore, a decrease in the transmittance of the Amide III vibration was more pronounced in the S3 samples than in the other samples. The S3 samples used only protein-rich egg albumin as the foaming agent. The decrease in Amide III was likely due to protein denaturation that occurred during foam mat drying at higher temperatures [30].



Figure 3. Fourier transform infrared (FTIR) spectroscopy observation result of sappanwood extract at different drying temperatures (40, 60, and 80 °C) and different foam formulations (A) S1, (B) S2, (C) S3, (D) S4, and (E) S5.

3.4. Moisture ratio observation

The moisture ratio of the sappanwood extracts at different foam formulations and drying temperatures are depicted in Figure 4. As seen in Figure 4, the moisture ratios of sappanwood extracts at the end of the experiment at a drying temperature of 80 °C were the lowest for all the drying formulations. At a higher drying temperature, the relative humidity of the drying air was lower, leading to the enhancement of the driving force (moisture content difference) for drying. As a result, the drying time was shorter [9].

As illustrated in Figure 4 parts B to E, the foaming agents significantly reduced the moisture ratio of the sappanwood extracts (S2–S5 samples). For example, at the same temperature (i.e., 60 °C), the S2 (Figure 4 part B) and S3 (Figure 4 part C) samples resulted in a moisture ratio of about 1.8–7.5 times lower than that of the S1 sample (Figure 4 part A) The foaming agents play an important role in the drying process by creating porous structures that result in a higher surface area required for moisture evaporation [10,11]. These porous structures can be clearly seen in the SEM images of the S2–S5 samples presented in Figures 2 parts B to E.

In this study, the S3 samples, which employed only egg albumin as the foaming agent, exhibited the largest and fastest moisture removal as compared to that of the others. The egg albumin contains a higher amount of protein (up to 12.5%) than that of the gum Arabic (up to 3%) [31,32], which was used as a substitute to egg albumin in the S2, S4, and S5 samples. The higher protein content in the S3 samples generated more stable foams for the sappanwood extract drying than those in S2, S4, and S5 samples. The addition of protein produces foam and improves the kinetic stability of the thermodynamically unstable foam. The ability of the egg albumin to stabilize the foam comes from the amphiphilic nature of the protein to emulsify the continuous and dispersion phases, which enhance foam flexibility [11]. This finding is in accordance with the SEM observations, wherein the porous structures in the S3 samples remained until the end of the drying process.

3.5. Drying kinetics model and predicted drying time

Four selected thin-layer drying models were fitted to the experiment foam mat drying data. For indicators, the coefficient of determination (\mathbb{R}^2) and the root-mean-square deviation (RMSD) values were evaluated. The result shows that all the thin-layer drying models resulted in a comparable \mathbb{R}^2 value (above 0.97). In this case, the Page model became the most favorable with lowest average RMSD of about 0.006 for drying temperatures ranging 40 °C to 80 °C. The drying rate constant (k) value, was ranged from 0.003 to 0.049 min⁻¹. While the n values were 0.697 to 1.697. This finding is in good agreement with previous reports that the Page model has been successfully used for describing agricultural product drying, such as for bay leaf [33], sweet cherry [34], and cassava starch [35].

Accordingly, the selected thin-layer drying model, namely, the Page model was then used to predict the drying time of the sappanwood extract using the foam mat drying process. Figure 5 displays the estimated drying time of five different foam formulations. The S3 formulation possesses the shortest drying time. It can be concluded that the addition of egg albumin as the foaming agent successfully sped up the drying process up to 3–7 times faster than the S1 formulation, in which no foaming agents were added. Egg albumin plays an important role in the formation of stable foams for sappanwood extract drying by enhancing foam stability [10,11]. Meanwhile, the gum Arabic can be used to decrease the stickiness of the sappanwood extract by increasing the glass transition

temperature [36]. By using a combination of gum Arabic and egg albumin, the drying process can be sped up, with a reasonable dry product quality.





The operational temperature also affects moisture reduction in the extract mixture. Higher drying temperature facilitates a more rapid drying process due to its higher driving force for the drying and requires a shorter drying time [9]. However, a higher drying temperature may lead to extensive degradation of active compounds in the sappanwood extract that will be discussed in the later section [29].



Figure 5. The estimated drying time at various drying temperatures (40, 60, and 80 °C) and foam formulations (S1, S2, S3, S4, and S5).

3.6. Total phenolic compounds

The total phenolic content of the sappanwood extracts obtained from the drying of five different foam formulations (S1, S2, S3, S4, and S5) at 40 °C–60 °C was evaluated using Folin–Ciocalteu colorimetric [17]. The total phenolic content of the sappanwood extracts obtained from drying at 80 °C was not tested, given that their color change and the OH group's reduction were the highest (Figure 3 parts A to E and Table 1).

During the drying process, the total phenolic compounds of sappanwood extract decreased with the increase in drying time. Figure 6 shows the thermal degradation of the total phenolic compounds during the drying process. A literature survey explained that several factors, including light, pH, oxygen level, and temperature, can affect the total phenolic compounds [20]. In the drying process, the sappanwood extract was in direct contact with hot drying air and oxygen. Consequently, heat and oxygen exposures lead to a reduction in the total phenolic compounds of the sappanwood extract.

Figure 6 presents that the S3 sample exhibited a higher reduction of total phenolic compounds during the drying process. The existence of protein contained in the foaming agents also induces the occurrence of the Maillard reaction, which is responsible for the degradation of the phenolic compounds. This observation could be the result of the binding between the phenolic compounds with proteins or by the structural changes of the phenolic compounds [37]. As expected, the addition of gum Arabic helped preserve the total phenolic compounds in the sappanwood extracts. With the introduction of heat, the gum Arabic created a more stable linkage at the surface of the sappanwood extracts, which prevented them from thermal degradation. The gum Arabic is a complex heteropolysaccharide that is usually used in the food industry because of the effectiveness to protect bioactive compounds from oxidation [35,36].

As seen in Table 2, the degradation of total phenolic compounds can be well-fitted by the firstorder kinetic model. Based on the R² values, which all were higher than 0.814, the proposed model successfully described the degradation of the total phenolic compounds in sappanwood extract drying. Table 2 confirms that the increase in drying temperatures leads to an increase in the degradation rate constant of the total phenolic compounds. In general, the degradation rate constants of the total phenolic compounds of the S3 samples were slightly higher than those of formulas without the addition of foaming agents (S1 samples). The results suggest that the addition of protein-rich foaming agents accelerates the degradation rate by up to 2.6 times.



Figure 6. Total phenolic compounds at different drying temperatures (A) 40 °C and (B) 60 °C and different foam formulations (S1, S2, S3, S4, and S5).

Foam formulation	T (°C)	$k \times 10^2$, min ⁻¹	R ²
S1	40	1.880	0.945
	60	0.960	0.908
S2	40	1.100	0.911
	60	0.540	0.841
S3	40	2.060	0.905
	60	2.510	0.814
S4	40	1.990	0.927
	60	0.530	0.875
S5	40	6.000	0.964
	60	0.730	0.902

Table 2. Kinetic parameters of total phenolic compounds degradation at 40 and 60 °C.

The percentage of the total phenolic compound degradation was calculated based on the value of the predicted drying time from Figure 7 and the total phenolic compound retention rate constant rate presented in Table 2. The percentage of the total phenolic compounds retention is presented in Figure 7. The addition of egg albumin in the S3 samples successfully sped up the drying time. However, the color changes and the degradation rate constant of the total phenolic compounds in the S3 samples were the highest. Figure 7 also exhibits that the highest total phenolic compounds retention in the S1 samples at a drying temperature of 40 °C was 18%. With the total phenolic compound degradation of 27%–29% or a retention value of 71%–73%, the preferred drying formulation was that of the S2 or S4 samples, with a drying temperature at 60 °C. This condition was then used for optimization.



Figure 7. The estimated percentage of total phenolic compounds retention.

3.7. Optimization of total phenolic compounds

The sappanwood extract was dried at various drying temperatures and foam formulations. The levels of the process variables were determined by the previous data above. From the previous data, it can be concluded that the higher drying temperatures, which were supposed to be 80 °C, resulted in the highest color change and OH group reduction. The preferred foam formulation was that of the S2 sample, wherein the ratio of egg albumin and gum Arabic was 2 (20% egg albumin, 10% gum Arabic), and that of the S4 sample, wherein the ratio of egg albumin and gum Arabic was 0.5 (10% egg albumin and 20% gum Arabic). Here the drying temperatures ranged from 40 °C to 60 °C, while for the foam formulation, the ratio of egg albumin (A) and gum Arabic (B) ranged from 0.5 to 2. The low value (-1), center value (0), high value (+1), and star point (± 1.414) of process variables are listed in Table 3. The optimization was conducted in 10 runs, and the experimental design used a central composite design (CCD).

Table 3. Rotatability central composite design for 2 variabels and observed response of total phenolic compounds retention.

Run	Ratio of Egg Albumin (A)	T (°C)	Total Phenolic Compounds Retention (%)
	and Gum Arabic (B)		
1	0.500 (-1)	40 (-1)	21.232
2	2 (+1)	40 (-1)	40.808
3	0.500 (-1)	60 (+1)	70.680
4	2 (+1)	60 (+1)	72.865
5	0.189 (-1.414)	50 (0)	43.703
6	2.311 (+1.414)	50 (0)	59.090
7	1.250 (0)	35.8 (-1.414)	22.581
8	1.250 (0)	64.1 (+1.414)	80.215
9	1.250 (0)	50 (0)	51.397
10	0.500 (-1)	40 (-1)	21.232

In this study, the process variables, namely, the ratio of egg albumin and gum Arabic (*A*) and drying temperature (*B*), and the response variables, namely, total phenolic compounds retention (*Y*), was expressed in a second-order polynomial model. Equation 8 was evaluated using the coefficient of determination (R^2) and analysis of variance (ANOVA). The value of R^2 of Equation 9 was 0.897, indicating good correlation between the experimental and predicted values.

$$Y = -18 + 22.8A - 0.25B + +5.36A^2 + 0.0302B^2 + -0.580AB$$
(9)

ANOVA was used to evaluate the significance of the second-order polynomial model. According to the ANOVA result, the temperatures significantly affected the total phenolic compound retention, with a p-value of 0.001 (p-value < 0.05 is significant).

Using Equation 9, the two- and three-dimensional plots of the total phenolic compound retention was obtained, as illustrated in Figures 8 parts A and B, respectively. The result indicated that at lower the ratio of egg albumin and gum Arabic, the total phenolic compound retention was increased. The maximum value of the retention was 87.25% or the degradation was 12.75%, which could be achieved by drying at 64.1 °C using a ratio of egg albumin and gum Arabic of 0.189 (5% egg albumin and 25%

gum Arabic) for 64.7 minutes. For comparison, for the blueberry extract, with maltodextrin and whey protein isolate as the foaming agents, was dried using a freeze dryer, the retention was 76%. In contrast, the introduction of heat by spray dryer at 150 °C caused total phenolic compound degradation of about 52% [40]. Furthermore, with a lower total phenolic compound degradation of the foam mat

sappanwood extract, this product can be an option for a natural food colorant that is very useful against



Figure 8. Two dimensional (A) and three-dimensional (B) plots of total phenolic compounds retention.

4. Conclusions

free radical attacks [2].

The sappanwood extract powder has been successfully dried using a foaming agent. The addition of only a protein-rich foaming agent (egg albumin 30%), called the S3 formulation, sped up the drying process up to 3-7 times faster than that without foaming agents. The SEM observations revealed that the addition of foaming agents could lead to the formation of stable foams that enlarge the surface area for moisture removal. The water removal can be well represented by the Page model having an R² close to 1 with the lowest RMSD of about 0.006. However, the presence of foaming agents also

promoted product browning due to the Maillard reaction, as indicated by product darkness. Consequently, the total phenolic content of the sappanwood extract was also reduced, as proven by the hydroxyl group peak reduction obtained from the FTIR analysis.—The addition of gum Arabic can inhibit the Maillard reaction, resulting in a better product appearance and total phenolic compound. However, the effect of drying time reduction is still inferior to that of the addition of egg albumin 30%. The combination of gum Arabic and egg albumin reveals better improvement both in drying time, product appearance, and total phenolic compound retention.

Meanwhile, for all cases, the increase in drying temperature drastically reduced the drying time. However, the products suffer from a significant decline in quality. Considering this trade-off, the optimization using the RSM was conducted. The results indicated that the most favorable condition for foam mat drying of sappanwood extract was drying at 64.1 °C using a mixture of 5% egg albumin and 25% gum Arabic as the foaming agent. With this drying condition, the drying time to obtain sappanwood extract dry powder was only 64.7 minutes with a total phenolic compound retention of about 87.25%.

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Conflict of interest

The authors declare no conflict of interest related to the work presented in this paper.

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