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Optimization of Spray Drying Process Parameter Condition for Theaflavin Extract from Black Tea (Camellia sinensis) Using Response Surface Methodology (RSM)

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***Abstract*— Black tea theaflavins contain high antioxidants that can be protective agents against SARS-CoV-2. The relatively good process conditions in spray drying are predicted to be able to protect and maintain the storage stability of theaflavin bioactive compounds by encapsulation. Therefore, this study aims to obtain optimum process conditions in spray drying black tea theaflavin extract. The analyzed data were interpreted using response surface methodology (RSM) with the independent variables used including spray drying inlet air temperature (100 ºC, 120 ºC, 140 ºC) and extract to maltodextrin ratio (1:0.40; 1:0.80; 1:1.20 (v/v)). The resulting black tea powder was analyzed for theaflavin content by UV-Vis spectrophotometer (380 nm), moisture content by moisture analyzer, particle size by scanning electron microscopy (SEM), and yield. The optimum condition was reached at an inlet air temperature of 116ºC and a ratio of black tea extract to maltodextrin 1:0.6627 (v/v) which resulted in R-square for yield, moisture content, and theaflavin content of 0.8946; 0.8016; 0.9324 respectively. The best theaflavin storage stability results were achieved in the seventh experiment with operating conditions of inlet air temperature 120ºC and extract to maltodextrin ratio 1:0.23431 (v/v), obtained theaflavin content ranged from 0.37125% - 0.52650% from day 0 to day 30.**

***Keywords*— Theaflavin; Black Tea; Spray Drying; Optimization; Response Surface Methodology**

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I. INTRODUCTION

Black tea theaflavin is a component resulting from the oxidation of the polyphenol group catechin compounds during the complete fermentation process of green tea leaves which gives the tea a brownish-yellow colour and a slightly astringent taste. Theaflavins have pharmacological properties because they are high in antioxidants that can be protective agents in a variety of diseases such as reducing the risk of cardiovascular disease, cancer, diabetes, SARS-CoV-2, and oral health [1], [2]. The theaflavin content of black tea is about 3-6% dry weight [3], [4]. However, theaflavin content in black tea is quite difficult to isolate because it easily undergoes thermal degradation into thearubigin compounds [5], [6].

Research related to theaflavin extraction has been intensively developed. Extraction of black tea theaflavins using Microwave Assisted Extraction (MAE) method with water solvent is an effective method to isolate black tea theaflavin compounds as it has higher energy efficiency level, less solvent used and fast extraction time [7]. However, black tea theaflavin extraction results tend to have low storage stability as theaflavin compounds in liquid form easily undergo chemical degradation with the length of storage time [6], [8]. Therefore, drying of black tea theaflavin extract is necessary in order to increase the durability of black tea theaflavin compounds during storage.

In general, tea extract drying techniques are carried out by freeze drying [9], [10]; particles from gas saturated solutions (PGSS) drying process [11]; and spray drying in the form of encapsulation that produces dry particles [12]. Freeze drying requires a long time, high energy consumption, low yield, and high operational costs [13]. Particles from gas saturated solutions (PGSS) drying process also takes a long time, and the equipment maintenance and operational costs are expensive [14]. Meanwhile, spray drying produces higher quality dried products than other drying equipment because it can maintain high-quality properties such as colour, taste, as well as nutrients, relatively short time used, low cost, and is very suitable for encapsulating materials containing thermally sensitive compounds [15]–[19]. Spray drying converts liquid feed into dry powder. The liquid feed is atomized and contacted with hot air resulting in evaporation into dry particles which will then be separated from the air stream [12], [20], [21]. Various studies related to spray drying have been successfully applied in drying bioactive compounds of tea extracts such as spray drying of black tea extract [22]; spray drying of mountain tea extract [23]; and spray drying of green tea extract [24].

The choice of wall material is also important for efficient spray drying [25]. Compare to various types of starch-derived wall materials, maltodextrin was found to be more effective in protecting important components such as aroma, color, product yield, and phenolic compounds in tea extracts due to its high solubility, low hygroscopicity and are affordable [24], [26], [27]. To find out the optimum process parameters, analysis software is needed that can predict and model the dependent variables effectively to find out the optimum process parameters. Response surface methodology (RSM) is an empirical statistical method used to analyze multiple regression and can be used in solving multivariable equations simultaneously using multivariable quantitative data. It is also widely used to examine the relationship between response and independent variables and is recognized as a comprehensive, concise, and highly efficient methodology [28]. Response surface analysis was used to obtain optimum process conditions in spray-drying black tea theaflavin extract. Various studies related to spray drying optimization with response surface methodology have been carried out, including spray drying of apple custard [29]; spray drying of kefir powder [30]; spray drying of lassi powder [31]; spray drying of red wine lees [32]; spray drying of green tea [12], [21]; and spray drying of spent black tea [28]. Hence, this study aims to determine the optimum operating conditions in spray drying black tea theaflavin extract using response surface methodology (RSM) with independent variables, including inlet air temperature and the ratio of theaflavin extract to carrier material to increase the stability of theaflavin products produced.

II. MATERIAL AND METHODS

A. *Material*

Crushing-Tearing-Curling (CTC) black tea was obtained from PT Perkebunan Nusantara VII (Lampung, Indonesia). CTC black tea is a tea that is blanched with a very vigorous grinding process to extract as much cell liquid as possible. Maltodextrin (MD12) was used as natural carrier agent, Aquadest as extraction solvent, ethyl acetate p.a and methanol p.a for the analysis of theaflavin content were purchased from Merck & Co, Inc (New Jersey, USA).

B. *Preparation of Black Tea Extraction*

Black tea was put in a flat bottom flask and added to distilled water with a tea: solvent ratio of 1:30. The black tea was then extracted using a Microwave (Microwave NeoChef^{TM} Smart Inverter) with 400 W power for 8 minutes. Furthermore, the black tea extract was removed, cooled, and filtered to separate the extract from the pulp. Fifty mL of black tea extract was then analyzed for theaflavin content using a UV-Visible Double beam spectrophotometer with a wavelength of 380 nm. The other extracts were added with maltodextrin (MD12) with extract: maltodextrin ratios of 1:0.40; 1:0.80; and 1:1.20 (v/v). After being homogenized at 1500 rpm (The RET basic Magnetic Stirrer, Basic Ika Labortechnik, Germany) for 30 minutes, the dispersion of black tea extract and maltodextrin was processed into powder using a Spray Dryer

C. *Spray Drying of Black Tea Extracts*

Spray drying was carried out with a laboratory-scale spray dryer (Büchi Mini Spray Dryer B-290, Switzerland) that has a two-fluid nozzle (0.5 mm inner diameter) and operates co-currently. The inlet temperature were varied at 100ºC, 120ºC, 140ºC, ± 80ºC outlet air temperature, 50% aspiration rate, and 10 ml/min feed flow rate. The resulting black tea extract of theaflavin powder was then placed in aluminum foil bags until further analysis.

D. *Analysis of Theaflavin Content of Black Tea Extract*

The method of analyzing theaflavins was previously described by Akuli [33]. Fifty ml of black tea extract sample was mixed with 50 ml of ethyl acetate in a separatory funnel. A blank solution was prepared by diluting 4 ml of ethyl acetate with 25 ml of methanol in a volumetric flask. The absorbance of the sample solution was then measured using a Double Beam UV-Visible Spectrophotometer at a wavelength of 380 nm. The theaflavin content was calculated using the following equation:
$\%\ \text{Theaflavin} = 2.25 \times E1$ (1)
Where E1 was the absorbance of solution A at a wavelength of 380 nm after standardizing with a blank.

E. *Moisture Content Analysis*

Moisture content analysis was carried out using a moisture analyzer (Mettler Toledo^{TM} HC 103 Halogen Moisture Analyzer), where 0.5 samples were weighed in triplicate and analyzed using a moisture analyzer until constant weight and then the results were recorded.

F. *Statistical Analysis*

Response Surface Methodology (RSM) was used to analyze optimization process between independent variables (inlet temperature (X_1) and ration extract : maltodextrin (X_2)) and response variables (yield, moisture content, theaflavin content day-0 (D0). Central Composite Design (CCD) Statistical Software (Minitab 19, Pennsylvania, US) was used for the experimental design. This design consisted of ten sets of experiments shown in **Table 1**.

TABLE 1.
EXPERIMENTAL DESIGN OF SPRAY DRYING PROCESS THEAFLAVIN BLACK TEA EXTRACT ACCORDING TO CENTRAL COMPOSITE DESIGN

Run	Inlet Air Temperature (℃)	Ratio Extract : Maltodextrin (v/v)
1	100	1:0,40
2	100	1:1,20
3	140	1:0,40
4	140	1:1,20
5	91,71573	1:0,80
6	148,28427	1:0,80
7	120	1:0,23431
8	120	1:1,36569
9(C)	120	1:0,80
10(C)	120	1:0,80

The experimental data from the design was analyzed by a nonlinear regression procedure using the following second-order polynomial equation :

$$Y = \beta_0 + \sum \beta_i X_i + \sum \beta_{ii} X_i^2 + \sum \beta_{ij} X_i X_j$$

Where Y is the predicted value of the response, β_0 is the intercept term, β_i is the linear term, β_{ii} is the quadratic term, β_{ij} the interaction term X_i and X_j is the code level of the independent variable. Lack of fit testing is used to evaluate the fit of the mathematical model. The Fischer value for lack of fit should not be significant for model fitting [34]. The optimum values of the independent variables were analyzed using the desirability function method. Analysis of variance (ANOVA) was used to generate a p-value that can determine the terms in the model are statistically significant.

G. Storage Stability

Black tea powder was stored in an incubator at 45°C for 30 days. Storage stability testing was conducted at specific periods (0, 15, and 30 days) to measure the stability of theaflavin content in black tea powder during storage.

H. Scanning Electron Microscopy (SEM) Analysis

Scanning Electron Microscopy (SEM JSM-6510LA) was used to analyze the particle size and morphological structure of spray-dried black tea extract powder at 250x, 500x, 1500x, and 5000x magnification levels.

III. RESULT AND DISCUSSION

A. Response Surface (RSM) Interaction Effect of Independent Variables on Physicochemical Properties of Theaflavin Powder Spray Drying Black Tea Extract

Table 2 shows the experimental design for percentage yield, moisture content, and theaflavin content of spray-dried black tea extract powder (Day-0). **Table 3** shows the p-value results of the analysis of variance (ANOVA) for percentage yield, moisture content, and theaflavin content of spray-dried black tea extract powder (Day-0) that can determine whether the terms in the model are statistically significant. A value of $p \leq 0.05$ indicates a significant relationship between the independent variable and the response. The insignificant Lack-of-Fit of all responses indicates that the polynomial model fits the experimental data well. This polynomial model was used to show the relationship between yield, moisture content, and theaflavin content respectively with the corresponding independent variables (inlet air temperature and ration extract: maltodextrin). As for the R^2 value (in the range of 0-1), the model should be more than 0.80 to show good prediction [29], [31]. In this case, the R^2 values were significantly matched for the yield, moisture content, and theaflavin models so that the models had a good ability to predict the responses (**Table 4**).

B. Parameter effect on yield

The yield of theaflavin powder spray drying black tea extract produced ranged from 4.94% - 12.11% in ten experiments. The lowest yield (4.94%) was obtained from the results of experiment seven at an inlet air temperature of 120ºC and an extract to maltodextrin ratio of 1:0.23431 (v/v). Meanwhile, the highest yield (12.11%) was obtained from experiment eight at an inlet air temperature of 120°C and extract to maltodextrin ratio of 1:1.36569 (v/v) **(Table 2)**. This response was calculated by the second-order polynomial equation for powder yield (Y_1) **(Table 4)**. **Table 4** also shows the coefficient of determination (R^2) of 0.8946 so the yield response has significance for this model. Analysis of Variance (ANOVA) was used to determine the significance and suitability of the experimental model **(Table 3)**. The linear effect of extract: maltodextrin ratio (X_2) showed significant results ($p \leq 0.05$) on powder yield (Y_1) as shown in Table 3. This is because increasing the concentration of maltodextrin carrier from extract: maltodextrin ratio of 1:0.23431 (v/v) at 120°C to 1:1.36569 (v/v) at 120°C resulted in a 7.17% increase in powder yield **(Table 2)**. An increase in yield with the addition of carrier was also reported for mountain tea extract spray drying powder with an increase in carrier concentration from 3 g/100 g to 5 g/100 g can increase about 36% powder yield which can be attributed to the higher solids content of the feed solution [23]. Similar results were also observed in the spray drying of dragon fruit extract [35] and sweet orange juice [36] where increasing maltodextrin concentration increased the process yield and vice versa. At lower maltodextrin concentrations, the feed has lower total solids so the viscosity of the feed is also lower. This allows for shear action between the high-velocity compressed air and the low-velocity feed resulting in high velocities in the atomized feed spray droplets so that the sprayed feed droplets collide with the internal walls of the drying chamber at a higher velocity and intensity and result in increased wall deposits which can decrease the process yield. The model F-value of 6.79 indicates that the model is significant, while the Lack of Fit F-value of 1.23 indicates the results are not significant to the pure error **(Table 3).**

TABLE 2.
EXPERIMENTAL DESIGN AND THE INFLUENCE OF INDEPENDENT VARIABLES ON THE RESPONSE OF CHANGES IN PHYSICOCHEMICAL PROPERTIES OF THEAFLAVIN POWDER FROM SPRAY DRYING OF BLACK TEA EXTRACTS.

Run	Inlet Air Temperature (℃)	Ratio Extract : Maltodextrin (v/v)	Product Yield (%)	Moisture Content (%)	Theaflavins Day-0 (D0) (%)
1	100	1:0,40	5,38	7,16	0,43200
2	100	1:1,20	8,48	7,21	0,04950
3	140	1:0,40	5,69	5,27	0,17775
4	140	1:1,20	8,91	6,26	0,04725
5	91,71573	1:0,80	7,58	6,04	0,07875
6	148,28427	1:0,80	6,41	4,31	0,08100
7	120	1:0,23431	4,94	7,35	0,37125
8	120	1:1,36569	12,11	5,90	0,06300
9(C)	120	1:0,80	7,96	5,28	0,09000
10(C)	120	1:0,80	6,61	4,92	0,13050

TABLE 3.
ANALYSIS OF VARIANCE (ANOVA) FOR YIELD, MOISTURE CONTENT, THEAFLAVIN

Source	DF	Y_1(%)			Y_2 (%)			Y_3 (%)		
		Adj SS	F-Value	P-Value	Adj SS	F-Value	P-Value	Adj SS	F-Value	P-Value
Model	5	36,3524	6,79	0,044	7,83682	3,23	0,139	0,000016	11,04	0,019
Linear	2	33,9707	15,86	0,013	3,62117	3,73	0,122	0,000012	20,84	0,008
X_1	1	0,1046	0,10	0,770	3,49350	7,21	0,055	0,000001	2,77	0,171
X_2	1	33,8661	31,62	0,005	0,12767	0,26	0,635	0,000011	38,90	0,003
Square	2	2,3781	1,11	0,414	3,99475	4,12	0,107	0,000002	4,01	0,111
X_1^2	1	0,4287	0,40	0,561	0,15018	0,31	0,608	0,000000	0,11	0,762
X_2^2	1	0,9621	0,90	0,397	3,75446	7,74	0,050	0,000002	5,78	0,074
2-Way Interaction	1	0,0036	0,00	0,957	0,22090	0,46	0,537	0,000002	5,49	0,079
X_1X_2	1	0,0036	0,00	0,957	0,22090	0,46	0,537	0,000002	5,49	0,079
Error	4	4,2840			1,93938			0,000001		
Lack-of-Fit	3	3,3728	1,23	0,566	1,87458	9,64	0,231	0,000001	4,37	0,335
Pure Error	1	0,9112			0,06480			0,000000		
Total	9	40,6364			9,77620			0,000017		

X_1 = Inlet Air Temperature, X_2 = Ration Extract : Maltodextrin, Y_1 = Yield, Y_2 = Moisture Content, Y_3= Theaflavin (Hari ke-0), DF = Degree of Freedom, Adj SS = Adjusted sums of squares, p-value = probability value ($p<0,05$)

TABLE 4.
REGRESSION EQUATION OF THE RESPONSE

Response	Response Quadratic polynomial model equations	Standard error	R^2
Y_1 =	$-5,0 + 0,175\ X_1 + 0,11\ X_2 - 0,00077\ X_1^2 + 2,87\ X_2^2 + 0,0037\ X_1\ X_2$	1.03490	0.8946
Y_2 =	$22,3 - 0,165\ X_1 - 12,90\ X_2 + 0,000453\ X_1^2 + 5,66\ X_2^2 + 0,0294\ X_1X_2$	0.696308	0.8016
Y_3 =	$0,01242 - 0,000030\ X_1 - 0,01846\ X_2 - 0,000000\ X_1^2 + 0,00378\ X_2^2 + 0,000079\ X_1X_2$	0.0005379	0.9324

X_1 = Inlet Air Temperature, X_2 = Ration Extract : Maltodextrin, Y_1 = Yield, Y_2 = Moisture Content, Y_3= Theaflavin Hari ke-0

C. Parameter effect on moisture content

The moisture content of theaflavin powder spray drying black tea extract produced was in the range of 4.31% - 7.35% in ten experiments. The lowest moisture content (4.31%) was obtained from the results of experiment six at an inlet air temperature of 148ºC and an extract-to-maltodextrin ratio of 1:0.80 (v/v). Meanwhile, the highest moisture content (7.35%) was obtained from experiment seven at an inlet air temperature of 120°C and an extract-to-maltodextrin ratio of 1:0.23431 (v/v) **(Table 2)**. This response was calculated by the second-order polynomial equation for moisture content (Y_2) **(Table 4). Table 4** also shows the coefficient of determination (R^2) of 0.8016 so the moisture content response has significance for this model. Analysis of Variance (ANOVA) was used to determine the significance and suitability of the experimental model **(Table 3).** The linear effect of inlet air temperature (X_1) showed a significant result ($p \leq 0.05$) on moisture content (Y_2) shown in **Table 3**. This is because an increase in inlet air temperature causes a decrease in moisture content. Similar results were reported in the spray drying of green tea extract [24]; acai powder [38]; watermelon juice [39] and Agaricus blazei murill polysaccharide [40] where the decrease in moisture content with increasing inlet air temperature is due to the higher inlet air temperature, the rate of heat transfer to the particles is greater, providing a greater driving force for the evaporation of moisture content so that a powder with low moisture content is formed. Research on the spray drying of green tea [21] also reported that very low drying inlet air temperature can result in higher moisture content in the product, causing the product to stick in the drying chamber, this can also affect the yield of the resulting product. Tengse [12] reported that increasing the drying inlet air temperature has decreased the relative humidity of the air which can take up more water vapor from the feed emulsion during spray drying, resulting in lower moisture content in the powder. In addition, the quadratic variable ration extract: maltodextrin x ration extract: maltodextrin (X_2^2) in the ANOVA results **(Table 3)** also showed significant results ($p \leq 0.05$) on moisture content (Y_2). This is because increasing the amount of maltodextrin carrier material can increase the total solids in the feed liquid and it will be difficult for water molecules to diffuse past large maltodextrin molecules because water molecules are retained in the matrix so that the moisture content can increase [36]. The model F-value of 3.23 indicates that the model is insignificant, while the Lack of Fit F-value of 9.64 indicates the results are also insignificant to the pure error **(Table 3).**

D. Parameter Effect on Theaflavin Content

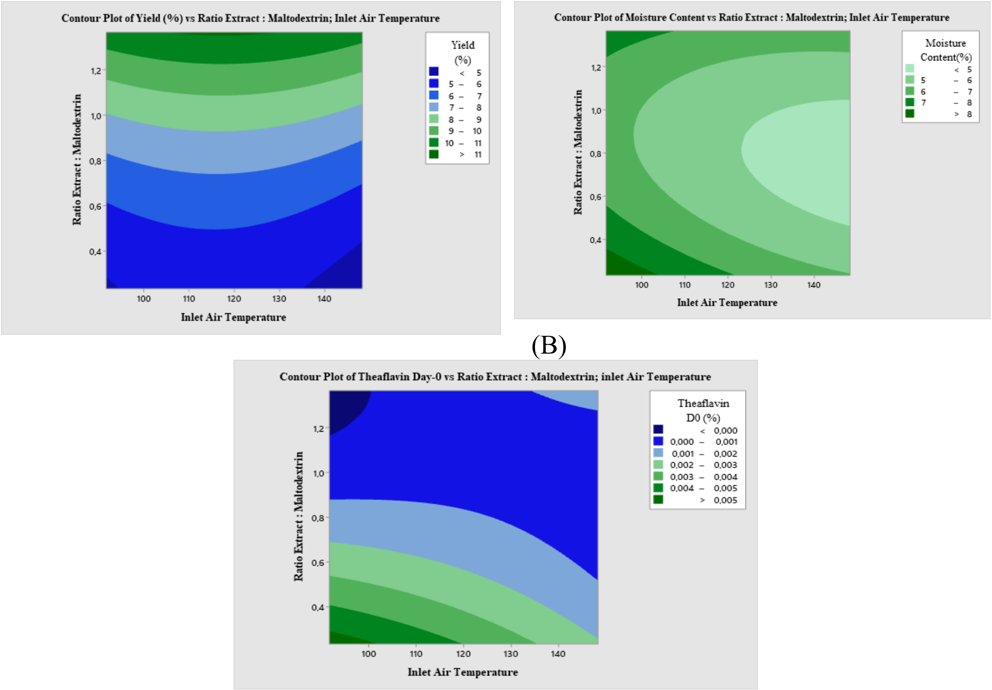
Theaflavin content of spray drying powder of day 0 black tea extract was in the range of 0.04725% - 0.43200% in ten experiments. The lowest theaflavin content (0.04725%) was obtained from the results of experiment four at an inlet air temperature of 140ºC and an extract-to-maltodextrin ratio of 1:0.20 (v/v). Meanwhile, the highest theaflavin content (0.43200%) was obtained from experiment one at an inlet air temperature of 100ºC and extract to maltodextrin ratio of 1:0.40 (v/v) **(Table 2).** This response was calculated by the second-order polynomial equation for theaflavin content (Y_3) **(Table 4). Table 4** also shows the coefficient of determination (R^2) of 0.9324 so the response of day 0 theaflavin content has good significance for this model. Analysis of Variance (ANOVA) was used to determine the significance and suitability of the experimental model **(Table 3).** The linear effect of extract: maltodextrin ratio (X_2) showed significant results ($p \leq 0.05$) on day 0 theaflavin content (Y_3) shown in **Table 3**. This is because high addition of maltodextrin can reduce theaflavin content. The same thing was confirmed in the spray drying study of mountain tea extract [23] which reported a decrease in total polyphenols of the sample with increasing concentration of maltodextrin carrier due to dilution of mountain tea extract with the addition of more carrier. The same was observed by Quek [39] for spray drying watermelon juice where if the percentage of maltodextrin was too high, the quality of the resulting powder would be lower as the nutrients from the watermelon juice would be dissolved and the red-orange color faded. Susantikarn & Donlao [24] also reported a decrease in polyphenol content in spray-drying green tea extract where an increase in maltodextrin carrier material resulted in low polyphenol content. The model F-value of 11.04 indicates that the model is significant, while the Lack of Fit F-value of 4.37 indicates the results are not significant to the pure error **(Table 3).**

E. Response of optimization of Yield, Moisture Content and Theaflavin at Day-0

The 3-D response surface contour plot **(Fig.1A)** shows the effect of inlet air temperature and ratio extract: maltodextrin on the powder yield response. The maximum yield of theaflavin powder spray drying black tea extract was obtained at the maximum extract: maltodextrin ratio of 1:1.36 (v/v) and inlet air temperature in the range of 110ºC - 130ºC.

Fig.1B is a contour plot of the effect of inlet air temperature and ratio extract: maltodextrin on the moisture content response of the powder. The results show that the minimum moisture content is obtained in the extract: maltodextrin ratio range of 1:0.45 - 1:1.0 (v/v) and inlet air temperature in the range of 125ºC - 148ºC.

Fig.1C shows the contour plot of the effect of inlet air temperature and ratio extract: maltodextrin on theaflavin content response of day 0 spray drying powder of black tea extract. The maximum theaflavin content was obtained at a minimum extract: maltodextrin ratio of 1:0.23 (v/v) and inlet air temperature in the range of 90ºC - 100ºC.



(A) (B)

(C)

Fig. 1 Contour Plot of the Effect of Independent Variables on the Response Surface for Yield (A), Moisture Content (B), and Theaflavin Content on Day-0 (C).

F. *Response Optimization*

The response optimization parameter of spray drying theaflavin of black tea extract made from maltodextrin wall over inlet air temperature and the ratio of black tea extract to maltodextrin were determined using minitab 19 software. The parameters to be obtained were to minimize moisture content, maximize theaflavin content and yield from ten running parameter designs (**Table 2**). The simulation aims to meet the good process condition in spray drying which can protect and maintain the storage stability of theaflavin bioactive compounds by encapsulation.

Table 5.
Parameters Response Optimization

Response	Goal	Lower	Target	Upper	Weight	Importance
Moisture Content (%)	Minimum		4,3100	7,35	1	1
Yield (%)	Maximum	4,94000	12,1100		1	1
Theaflavin D0 (%)	Maximum	0,00047	0,0043		1	1

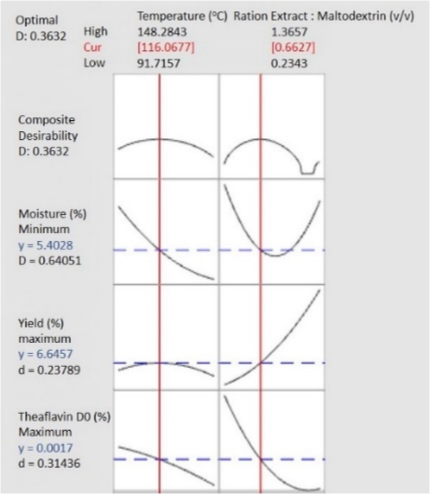


Fig. 2 Multiple Response Prediction on the Minimum Value of Moisture Contents, Maximum Value of Yield and Theaflavin Contents on Day-0 (C).

Based on the results shown in **Table 2**, the target theaflavin content, moisture content, and yield to be obtained were 0.0043%; 4.31%; and 12.11%; respectively. Therefore, the solution for optimum condition of the spray drying process of black tea extract was shown in **Fig.2**, with an inlet air temperature of 116°C and an extract : maltodextrin ratio of 1:0.6627. The responses fit the target values, with theaflavin content, moisture content, and yield obtained were 0.0017%; 5.4028; and 6,6457; respectively. Whereas the desirability value obtained was 0.3632. Desirability value indicates that the combination of simultaneous independent parameter levels fulfills the response requirements in the design. If the desirability value is closer to 1.0, it indicates the parameter condition to produce a more perfect product as expected. Despite the result of desirability value in this research was not closer to 1, a target in the particular responses highlights achievement in a particular aspect of the process or product which provides a basis for further research as a continuous improvement.

G. Storage Stability Analysis of Theaflavin Content

Table 6 shows the results of theaflavin levels before and after spray drying and the storage stability of theaflavin powder for 30 days. Theaflavin content in black tea extract before spray drying was 4.02469% and tended to decrease during the spray drying process because theaflavin compounds are sensitive to hot temperatures so they easily undergo chemical degradation during the drying process. The same thing was reported for spray drying green tea where polyphenols are sensitive to high temperatures, which also causes discoloration of the powdered product [21]. The effect of storage temperature and thermal processes on catechins, procyanidins, and total flavonoids of cocoa powder, that there are several polyphenolic compounds, namely flavonoids, catechins, and procyanidins that are sensitive to heat and can undergo chemical degradation during the heating process or even storage [5]. In addition, spray drying of green tea extract also reported a decrease in total polyphenol content as the inlet air temperature increased due to the loss of phenolics through the exhaust of the spray dryer during the drying process [12]. The total polyphenol content was higher at lower temperatures and higher concentration of green tea extract which can be observed in the ratio of green tea extract: 1:2 dressing with 25% green tea extract concentration and 120ºC temperature, the total polyphenol content was 57.81 mg/g dry matter.

During the storage stability test, the average theaflavin content tended to increase (did not decrease during storage). The highest theaflavin content was in the operating conditions of an inlet air temperature of 120ºC and the ratio of extract to maltodextrin of 1:0.585786, where theaflavin content ranged from 0.37125% - 0.52650% from day 0 to day 30. However, there was a decrease in theaflavin levels in sample-1 where theaflavin levels on day 0 decreased and then stabilized in storage from day 15 to day 30. The increase in theaflavin levels in the "dust" type of black tea occurred because the ability of oxygen was stronger so the oxygen reaction took place faster [41]. Oxygen will oxidize polyphenols into theaflavins and thearubigins in black tea during storage. Therefore, the polyphenol content in black tea powder is oxidized to theaflavin compounds which causes theaflavin content in black tea extract powder to increase during storage. While in sample-1 day 0 theaflavin content decreased on day 15 because some of the theaflavin content began to oxidize into thearubigin compounds. In the study of spray-drying of cranberry juice, the storage stability of polyphenol content (TPC) was analyzed from time 0 – 12 weeks [42]. At 0 – 6 weeks, the TPC was relatively consistent with some fluctuations in certain samples. During 6 – 12 weeks, powder TPC increased with storage time and reached the highest retention at week 8. This increase reached a 140% increase in TPC due to the presence of newly produced polyphenolic compounds during storage time. Similarly, the study of spray drying of grape pomace found an increase in the retention of polyphenols in encapsulated grape pomace during storage, due to the hydrolysis of conjugated polyphenols during storage, the polyphenol content changes as some compounds may degrade while other new polyphenols may form [43].

TABLE 6.
STABILITY STORAGES OF THEAFLAVIN CONTENT OF SPRAY DRYING BLACK TEA EXTRACT POWDER

Run	**Inlet Air Temperature (ºC)**	**Ratio Extract : Maltodextrin**	**Theaflavin Content**			
			Black Tea Extract (g/ml)	**Day-0 (D0) (%)**	**Day-15 (D15) (%)**	**Day-30 (D30) (%)**
1	100	1:0,40	4,02469	0,43200	0,32400	0,32175
2	100	1:1,20	4,02469	0,04950	0,06300	0,09675
3	140	1:0,40	4,02469	0,17775	0,25200	0,32850
4	140	1:1,20	4,02469	0,04725	0,06300	0,11700
5	91,71573	1:0,80	4,02469	0,07875	0,09450	0,18000
6	148,28427	1:0,80	4,02469	0,08100	0,13950	0,18225
7	120	1:0,23431	4,02469	0,37125	0,48375	0,52650
8	120	1:1,36569	4,02469	0,06300	0,07200	0,09675
9(C)	120	1:0,80	4,02469	0,09000	0,10575	0,12150
10(C)	120	1:0,80	4,02469	0,13050	0,13950	0,15525

D0 is Day-0 theflavin powder; D15 is Day-15 theflavin powder; and D30 is Day-30 theflavin powder

In addition, the addition of maltodextrin to spray-drying black tea extract can protect theaflavin compounds and increase storage stability so that they do not experience chemical degradation (decrease in levels) during storage. Several studies have also reported that the use of maltodextrin during the drying process can increase the shelf life of various compounds such as flavonoids, polyphenols, theaflavins, and catechins [44], [45]. In addition, research on microencapsulation of polyphenolic compounds from the red grape pulp [32] and spray drying of grape skin [46] reported that polyphenols dried by spray drying showed improved stability compared to alternative storage methods (freeze-drying or simple aqueous extraction) due to the protective effect of the carrier material.

H. Scanning Electron Microscopy (SEM)

The morphological properties of black tea extract theaflavin powder particles were analyzed using Scanning Electron Microscopy (SEM) **(Fig.3).** The SEM results showed the spray drying powder particles of black tea extract were 1-10 µm in size. Visually, theaflavin extract black tea powder particles were irregularly round and dented or concave without cracks on the surface. Similarly, it was observed in the spray drying study of green tea extract [21] that the resulting powder particles showed a relatively smooth structure without any cracks or fissures and some particles had a deformed structure on the surface due to heat, friction, rapid evaporation of water, or insufficient concentration for complete encapsulation of the tea extract. Meanwhile, the puckered and concave outer surface is characteristic of all powders dried by spray drying.

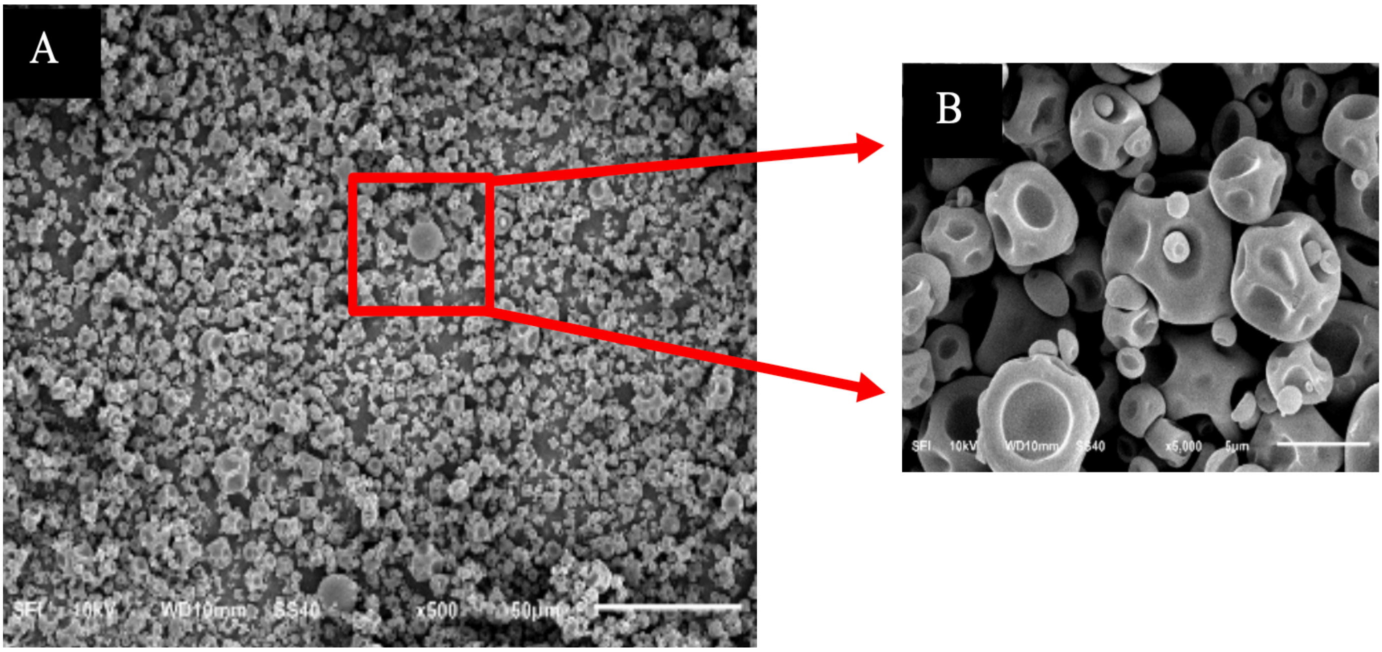


Fig. 3 Scanning Electron Microscopy (SEM) images of spray drying powder of black tea theaflavin extract at 500x (A) and large image at 5000x (B) magnification.

CONCLUSION

Experimental design using response surface methodology (RSM) central composite design (CCD) was conducted to analyze the effect of drying inlet air temperature and the ratio of black tea extract to maltodextrin carrier on the yield, moisture content, and theaflavin content of encapsulated black tea extract powder. The second-order polynomial model was found to be suitable for predicting the response model between independent variables. Optimum conditions were reached at an inlet air temperature of 116ºC and a ratio of black tea extract to maltodextrin of 1:0.6627 (v/v) which resulted in R-square for yield, moisture content and theaflavin content of 0.8946; 0.8016; 0.9324, respectively. The best theaflavin storage stability results were achieved in the seventh experiment with operating conditions of 120ºC inlet air temperature and the ratio of black tea extract to maltodextrin carrier 1:0.23431 (v/v), obtained theaflavin content ranging from 0.37125% - 0.52650% from day 0 to day 30. Therefore, the optimized black tea extract theaflavin spray drying powder can be included in foods to improve the nutritional value of foods

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CONFLICT OF INTEREST

The authors declare no conflicts of interest or personal relationships with other people or organizations that can inappropriately influence this work.

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