DEVELOPMENT OF MICROENCAPSULATED BIOACTIVE PRODUCT FROM *M. stenopetala* LEAVES EXTRACT BY OPTIMIZING SPRAY DRYING PROCESS PARAMETERS

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Abstract
Moringa stenopetala is a native plant in Ethiopia, which has a potential health effects due to its high bioactive contents. The aim of this study was to optimize spray drying microencapsulation conditions to maximize the encapsulation efficiency and total phenolic contents from M. stenopetala leaf extract. A Box-Behnken design was used to evaluate the influence of a spray drying process conditions such as maltodextrin: high methoxyl pectin (MD:HP) ratios (10:0, 9.5:0.5, and 9:1), core: coating ratios (1:10, 1:8, and 1:6), and inlet air temperature (120, 140 and 160 °C) on the encapsulation efficiency and total phenolic content (TPC) of microencapsulated bioactive products. The result showed that these parameters had significant effects on the responses. The encapsulation efficiency and the total phenolic contents of the microencapsulated bioactive product were in the range of 82.07–89.22% and 73.87–106.13 mg GAE/g micro, respectively. The spray drying process conditions considerably affected the physical and functional properties of the microencapsulated bioactive product. Similar trends were also observed on the antioxidant activities of the microencapsulates. The best encapsulation efficiency and TPC were obtained at the optimum spray drying process conditions: MD:HP ratio, core: coating ratio and inlet air temperature were: 9:1, 1:6 and 140 °C, respectively; with the predictive values of 87.8% for encapsulation efficiency and 103.55 mg GAE/g micro for TPC.

Keywords: *Moringa stenopetala*, spray drying, microencapsulation, optimization, bioactive product

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1. INTRODUCTION

Bioactive compounds are secondary metabolites of the plant, which are produced to increase their overall ability to survive and withstand challenges by allowing them to interact with their surroundings (Bernhoft, 2010). The epidemiological study shows that consumption of these vegetables decreases the risk of many diseases including different type cancers (Kim and Milner, 2005). As reported by Pappa et al. (2006), the *in vivo* and *in vitro* studies showed that among the vegetable the genus brassica has gotten more attentions because of their anticancer activity. This includes Moringa, due to the presence of bioactive compounds including enzymatic degraded products of glucosinolates (Bennett et al., 2003). Furthermore, the phenolic and flavonoid content are the major bioactive constituents of Moringa leaves that exhibit antioxidant activities (Habtemariam and Varghese, 2015; Vongsak et al., 2013a). The higher rutin content was found in *M. stenopetala* than Indian *M. olifiera* (Habtemariam and Varghese, 2015). Due to these reasons, the importance of these bioactive compounds and their possible use in processed foods as a natural antioxidant has been increasing.

The dried products have higher storage stability besides it is an advantage for ease of packaging and transportation due to the lower mass and volume (Caliskan and Dirim, 2013). However, these bioactive compounds are significantly reduced during conventional thermal processing (Abuye et al., 2003) and storage (Vongsak et al., 2013b). As indicated by Arun et al., (2011), one-fourth of the moringa was spoiled before consumption and 40% antioxidant activity was reduced during drying (Wangcharoen and Gomolmanee, 2013). In the

other vegetables, more than 50% of the glucosinolate lost during cabbage boiling, 23% of the antioxidant activity lost during cauliflower blanching (Puupponen-Pimiä et al., 2003), and 14% reduction of oxygen radical absorbance capacity of broccoli after it had been cooked. Significant loss of vitamin C by leaching and thermal effects has been also reported (Davey et al., 2000). These indicate that bioactive compounds are highly susceptible to the environmental factors.

Therefore, encapsulation can be as an alternative method to improve the stability of bioactive compounds during processing and storage. Microencapsulation is a technology for packaging of the active ingredients in micro particle matrices that forms the physical barrier between the active compounds and the external environment and to control the release of active compounds (Zuidam and Shimoni, 2010; Sagis, 2015). It also masks the bitter taste and odor [14]. Nowadays, microencapsulation has turned from simple encapsulation to the smart encapsulation with control release of the active compounds (Sagis, 2015).

Although there are numerous coating materials from proteins, modified and native starch, the commonly used is maltodextrin. This is due to its ability to form a film, high water solubility property, lower viscosity at higher concentration and lower cost (Sagis, 2015; Sansone et al., 2011). However, the efficiency and stability of the microencapsulate was improved when maltodextrin was mixed with other coating materials (Saénz et al., 2009; Tolun et al., 2016). Pectin is a biodegradable polysaccharide that is used in the food industry as thickening, stabilizing, gelling and film forming agent (Sagis, 2015; Sansone et al., 2011). In addition, it can be applicable in pharmaceutical industries as coating material for the control releasing system (Sagis, 2015).

There are different types of microencapsulation technology with their own advantages and disadvantages. Among these, spray-drying is the most commonly used technologies. Even if spray-drying processing destructs the heat sensitive and volatile compounds, it is a widely used technique, economical and applicable at large scale (Gharsallaoui et al., 2007). However, as to our knowledge, no published research was found on the development of microencapsulated bioactive product from *M. stenopetala* leaves extract by optimizing spray drying processing conditions. Therefore, the aim of this study was to optimize spray drying microencapsulation conditions for the development of bioactive product from *M. stenopetala* leaves extract and to evaluate the physical properties, bioactive compounds and antioxidant activity.

2. MATERIALS AND METHODS

Sample Collection

The *M. stenopetala* leaf sample was collected from Arba Minch, located at 6°01'59" N and 37° 32'59" E, at a distance 505 km from the capital city, Addis Ababa, Ethiopia. The collected sample was washed immediately using distilled water to remove dirt. Subsequently, it was dried for 72 h in a room with an average temperature of 25 °C and 62% of relative humidity. The dried sample was ground and allowed to pass through a sieve (20 meshes), kept in a sealed polyethylene bag, and stored in the dark until extraction was done.

Extraction of Bioactive Compounds

The extraction was done according to Vongsak et al. (2013a) with some modifications. The sample was mixed with 70% ethanol in the ratio of 1:40 (m/V). The extraction was done using the shaking water bath (JSSB-50T, JS Research Inc., Gongju, Korea) at a temperature of 30 °C for 24 h. Then, the extract was filtered using the Whatman No. 1 filter paper and dried using a rotary evaporator (Eyela, N-11, Tokyo, Japan) under vacuum at a temperature of 40 °C. The dried extract was stored in the refrigerator at 4 °C.

Spray Drying Encapsulation

Maltodextrin (Cornstarch, Eisse Food Co. Ltd., Korea) with a dextrose equivalent (DE) of 14–20, and high methoxyl pectin (Pectin RS400, Danisco Inc., USA) were used as encapsulating materials. Maltodextrin (MD) solutions were

prepared by mixing with distilled water in proportions of 10, 9.5 and 9% (w/V) using a magnetic stirrer. Pectin (HP) was added to MD solution to obtain total solid content of 10% (w/V) with the ratios of 10:0, 9.5:0.5 and 9:1 (w/w). The mixture was mixed using a magnetic stirrer. Then after, the extract was then mixed with core:coating ratio of 1:10, 1:8 and 1:6 (w/w). Then, the mixtures were homogenized using a high-speed homogenizer (T 25 ULTRA-TURRAX, IKA, Germany) at 35000 rpm for 3 min. The solutions were then put on the ultrasonicator for 5 min.

The spray drying was done according to the method described by Sansone et al. (2011) with some modifications. Lab scale spray dryer (Lab Plant, SD-06, Hunmanby, England) was used, which has two fluid nozzles with standard 0.5 mm jet. The spray dryer was adjusted to the conditions: air speed exhaust was 4.3 m/s, liquid flow rate was 485 mL/h, the deblocker was set at medium and a compressor of two bars. The microencapsulation was done at inlet temperatures of 120, 140 and 160 °C. The mixture was continuously stirred during spray drying was performed. Finally, the dried sample was packed and stored in a desiccator for further analysis.

Moisture Content

The moisture content was determined according to AOAC (2000). About 2 g of the sample was put in the pre-weighed aluminum can and kept in the drying oven (FO-600M, Jeion Tech, Korea) at 105 ± 1 °C for 6 h. The dried weight was measured and the percentage of the moisture content was determined.

Bulk Density

The bulk and tapped density were determined according to Deore et al. (2013) with minor modifications. About 5 g of the sample was put in a 25 mL graduated cylinder. Then, the occupied volume was recorded. The tapped density was determined after the graduated cylinder was tapped from about 5 cm height until constant volume was obtained. The volume was also recorded. Finally, the density expressed by dividing the weight of the sample by the occupied volume.

Water Absorption Index and Water Solubility Capacity

Water absorption index (WAI) was determined according to Paini et al. (2015) with minor modifications. Briefly, about 1 g of the sample was taken and placed in the pre-weighed 50 mL of centrifuge flask. It was put in 12 mL distilled water and mixed vigorously followed by agitation in the shaking water bath at 30 °C for 30 min. Then, the mixture was centrifuged at 1915 x*g* for 15 min. The supernatant was collected from the hydrated gel. Then, the hydrated gel was weighed and the WAI was calculated using Eq. 1.

$$WAI = \frac{Weight\ of\ hydrated\ gel}{Weight\ of\ sample} \qquad Eq.\ 1$$

To determine the water solubility capacity (WSC) of the microencapsulate, the supernatant was transferred to the pre-weighed aluminum can. Then, it was kept in the drying oven at 105 °C overnight. Finally, WSC was calculated using Eq. 2.

$$WSC\ (\%) = \frac{Weight\ of\ dried\ supernatant}{Weight\ of\ sample} * 100 \qquad Eq.\ 2$$

Hygroscopicity

The hygroscopicity was determined according to the method described by Gallo et al. (2015) with minor modification. About 1 g of the sample was put on the pre-weighed aluminum can. The sample was kept in airtight desiccator containing a saturated sodium chloride solution with the relative humidity of 75%. After one week, the sample was weighed and the hygroscopicity of the sample was calculated using Eq. 3.

$$Hygroscopicity\ (\%) = \frac{W_2 - W_1}{W_1} * 100 \qquad Eq.\ 3$$

Where, W_1 is the sample weight before treatment (g) and W_2 is the sample weight after treatment (g)

Encapsulation Efficiency

The encapsulation efficiency was evaluated using the method described by Mahdavi et al. (2016). To determine the total phenolic content, 100 mg of the microencapsulate was mixed with 1 mL of distilled water and stirred to dissolve thoroughly. Then, 9 mL of ethanol was added and mixed again for 5 min. The solution was filtered through a membrane filter with a pore size of 0.45µm. On the other hand, to extract the surface phenolic compounds from microencapsulate wall, 100 mg of the microencapsulate powder was taken and mixed with 10 mL of ethanol. The mixture was vortexed for 10 s and centrifuged at 1915 x*g* for 3 min. Finally, the clear supernatant was collected and filtered using 0.45µm membrane filter. Then, the total and surface phenolic compounds was quantified. Then, the encapsulation efficiency was determined using Eq. 4.

$$EE\ (\%) = \frac{(TPC - SPC)}{TPC} * 100 \qquad Eq.\ 4$$

Where, *EE* is encapsulation efficiency, *TPC* is the total phenolic content, *SPC* is the surface phenolic contents.

Total Phenolic Content

The total phenolic content (TPC) was measured according to Singleton and Rossi (1965). Briefly, 0.5 mL of the product (1 mg/mL) was mixed with 2.5 mL of 10% Folin-Ciocalteu reagent and vortexed. After 8 min, 2 mL of 7.5% sodium carbonate was added, mixed and kept in the dark at room temperature for 2 h. The same procedure was used for the blank and gallic acid standard at different concentrations to develop a standard curve. The absorbance was measured at 765 nm using a UV-Vis spectrophotometer (Optizen 2120 UV, Mecasys Co., Ltd, Korea). Then, the TPC was expressed as mg of gallic acid equivalent per gram of microencapsulates (mg GAE/g micro).

2, 2-diphenyl-1-picrylhydrazyl radical Scavenging Assay

The 2,2-diphenyl-1-picrylhydrazyl (DPPH) free radical scavenging activity was measured using the procedure described by Brand-Williams et al. (1995). Briefly, 2.4 mg of DPPH solution was prepared in 100 mL of 80% methanol and the absorbance was checked for the reading less than one at 515 nm. This was used to ensure the concentration of the reagent is at optimum for the determination of the scavenging activity. Then, 0.1 mL of the sample was mixed with 3.9 mL of diluted DPPH solution. The mixture was then mixed and kept in the dark at room temperature for 30 min. The same procedure was used for the Trolox standard of different concentrations and blank. Then, the absorbance was measured using a UV-Vis spectrophotometer at 515 nm. Finally, the antioxidant activity was calculated from the Trolox standard curve and expressed as mg Trolox equivalent per gram of microencapsulates (mg TE/g micro).

Ferric Reducing Power Assay

The ferric reducing antioxidant power (FRAP) assay was evaluated according to the method described by Nguyen et al. (2015). First, FRAP solution was prepared by mixing 300 mM acetate buffer solution (pH=3.6), 10 mM TPTZ solution in 40 mM hydrochloric acid and 20 mM ferric chloride solution in the ratio of 10:1:1; respectively. Then, 0.15 mL of the sample was mixed with 2.85 mL of fresh FRAP solution. The mixture was then kept for 30 min in the dark at ambient temperature. The same procedure was used for the blank and Trolox standard at different concentration. Then, the absorbance was measured using a UV-VIS spectrophotometer at 593 nm. Reducing power was calculated from the Trolox standard curve and expressed as mg of Trolox equivalent per gram of microencapsulates (mg TE/g micro).

Ferrous Ion Chelating Activity Assay

This chelating activity of the extract was done based on the procedure described by Chew et al. (2009). Briefly, 1 mL of 0.1 mM ferrous sulfate was added to 1 mL of the extract followed by adding 1 mL of 0.25 mM ferrozine solution. The mixture was shaken vigorously and allowed to stand for 10 min at room

temperature in the dark. The same procedure was done for EDTA standard with different concentrations and for the blank. Then, the absorbance was measured using a UV-Vis spectrophotometer at 562 nm. The ferrous ion chelating activity of the sample was calculated and described as mg EDTA equivalent per gram of the microencapsulates using the EDTA standard curve (mg EE/ g micro).

Color Analysis

Color measurement was done using a color analyzer (CR-400, Chroma meter, Konica Minolta, Japan) according to CIE (L^*, a^*, b^*) system. The CIE L^* (whiteness/darkness), a^* (redness/ greenness), b^* (yellowness/ blueness) were measured, and the chroma values (C^*) an hue angle (H^*) were calculated using Eq. 5 and Eq. 6, respectively.

$$C^* = (a^{*2} + b^{*2})^{1/2} \qquad Eq.\ 5$$

$$H^* = tan^{-1}(b^*/a^*) \qquad Eq.\ 6$$

Experimental Design and Statistical Analysis

or the optimization of spray drying process conditions of microencapsulation, the experimental results of the response surface design were analyzed using Design Expert software (State-Ease Inc., Version 7, Minneapolis, MN, USA,). Three independent variables such as maltodextrin:pectin ratio (A, %), core:coating ratio (B, %) and inlet temperature (C, °C) were used to analyze their effects on dependent variables such as encapsulation efficiency and total phenolic content (TPC), using Box-Behnken Design. The complete design consists of 17 experimental points (Table 1).

Table 1. Experimental design with the observed responses of encapsulation efficiency and TPC of the microencapsulated bioactive compound from *M. stenopetala* leaves extract

	A	B	C	Responses Y	
Run	MD:HP ratio (g/g)	Core:coating ratio (g/g)	Inlet temperature (°C)	Encapsulation efficiency (%)	TPC (mg GAE/g micro)
1	10:0	1:10	140	89.22	85.77
2	10:0	1:8	160	85.48	87.61
3	10:0	1:8	120	82.07	104.32
4	10:0	1:6	140	83.52	97.34
5	9.5:0.5	1:10	160	88.30	73.87
6	9.5:0.5	1:10	120	84.95	74.61
7	9.5:0.5	1:8	140	85.27	91.86
8	9.5:0.5	1:6	160	82.76	75.00
9	9.5:0.5	1:6	120	84.68	106.13
10	9:1	1:10	140	89.02	77.59
11	9:1	1:8	160	85.16	89.27
12	9:1	1:6	140	87.93	102.98
13	9:1	1:8	120	87.03	105.28
14	9.5:0.5	1:8	140	85.18	91.54
15	9.5:0.5	1:8	140	85.45	93.24
16	9.5:0.5	1:8	140	85.18	92.02
17	9.5:0.5	1:8	140	85.32	90.79

TPC= Total phenolic content; MD:HP= maltodextrin and high methoxyl pectin ratio

Subsequently, the output of encapsulation efficiency and TPC were measured from the microencapsulates. Then, it was optimized simultaneously by using respond surface method. The actual values used in the response surface analysis and the corresponding parameter values were shown in Table 1. A quadratic polynomial equation was selected to model the treatment effects and treatment interactions that were used to predict the optimal extraction parameters.

$$Y = \beta_o + \beta_1 A + \beta_2 B + \beta_3 C + \beta_{12} AB + \beta_{13} AC + \beta_{23} BC + \beta_{11} A^2 + \beta_{22} B^2 + \beta_{33} C^2 \quad Eq.\ 7$$

Where Y is the response variable; A, B and C are the independent variables for the maltodextrin:pectin ratio, core:coating ratio and inlet air temperature, respectively; β_0 is constant; β_1, β_2, β_3 are linear coefficients; β_{12}, β_{13}, β_{23} are interaction coefficients and β_{11}, β_{22}, β_{33} are quadratic coefficients for maltodextrin:pectin ratio, core:coating ratio and inlet temperature, respectively.

To compare other physical and functional properties, the bioactive content and antioxidant activities of the microencapsulated bioactive product, means comparison were done using one way analysis of variance (ANOVA), Tukey HSD test using JMP software (Version 13.0, 2016, SAS institute Inc., Cary, NC, USA). All experiments were conducted in triplicate and expressed as mean ± standard deviation. The difference was considered at a significance level of $P \leq 0.05$.

3. RESULTS AND DISCUSSION

Fitting Response Surface Model

The spray-drying process conditions to encapsulate the bioactive compounds of *M. stenopetala* leaves extract were optimized based on the encapsulation efficiency and total phenolic content with different maltodextrin: pectin ratio, core: coating ratio, and inlet air temperature as processing conditions. To set the range of parameters to be optimized, the preliminary experiments were done (data not shown). The pectin as a carrier agent was put as maximum 1% to minimize the gelling property. The inlet air temperature set to the minimum 120 °C to minimize insufficient moisture removal. This showed that the processing parameters had effects on encapsulation efficiency and total phenolic of the microencapsulated product.

The response surface model was developed using spray-drying process conditions to predict the encapsulation efficiency and total phenolic content (TPC) of the microencapsulated bioactive product from *M. stenopetala* leaf extract. The determination coefficient (R^2) of encapsulation efficiency was 99.7%, whereas 99.51% for TPC, which were closer to the adjusted R^2 of 99.33% and 98.89%, respectively. In addition, the predicted R^2 ranged from 96.39% to 94.71%, which shows reasonable agreement with the adjusted R^2 (Table 2). This showed that the quadratic model was adequately fitted to the encapsulation efficiency and TPC. The ANOVA results for the response surface quadratic model developed using the spray drying process parameters for encapsulation efficiency and TPC are shown in Table 2.

The *F* values of the models were 262.60 for encapsulation efficiency and 159.39 for TPC. These indicate that the models are highly significant ($P < 0.0001$) for the encapsulation and TPC. According to this study, the parameters, except A and AC interaction, had significant effects (Table 2). The lack of fitness was not significant ($P > 0.05$), which indicates the fitness of the polynomial model to the experimental values.

Effects of Spray Drying Process Conditions on Encapsulation Efficiency

The encapsulation efficiency is an important parameter for the microencapsulates, which determine the stability and control release of bioactive compounds. The spray drying parameters are the source of the differences for the effectiveness of encapsulation (Saikia et al., 2015).

Table 2. Regression coefficients for the fitted quadratic polynomial model and analysis of variance for the experimental results of encapsulation efficiency and TPC

Factors	Encapsulation efficiency (%)	TPC (mg GAE/g micro)
Intercept	85.28	91.89
Linear		
A- MD:HP ratio	-1.10[a]	-0.01[ns]
B- Core: coating ratio	1.58[a]	-8.70[a]
C- inlet air temperature	0.37[a]	-8.07[a]
Interaction		
AB	1.15[b]	3.45[b]
AC	1.32[a]	-0.18[ns]
BC	1.32[a]	7.60[a]
Quadratic		
A^2	0.95[a]	6.62[a]
B^2	1.19[a]	-7.59[a]
C^2	-1.30[a]	-1.89[c]
R^2	0.9970	0.9951
Adj. R^2	0.9933	0.9889
Pred. R^2	0.9639	0.9471
P-value	<0.0001	<0.0001
F-value	262.60	159.39
C.V. %	0.20	1.25
F-value Lack of fit	3.77	2.43

TPC- Total phenolic content; [a] Significant at P ≤ 0.0001; [b] Significant at P ≤ 0.001; [c] Significant at P ≤ 0.05; [d] and [ns] Non significant

According to this study, the core:coating ratio had significant effects on encapsulation efficiency (Fig. 1a). The core:coating ratio (1:10) gave higher encapsulation efficiency when it was compared to 1:6 and 1:8 ratios (Fig.1). This might be due to the presence of more surface phenolic compounds on the microencapsulates coated with high core:coating ratio, which was with the core coating ratio of 1:6. On the contrary, the lower surface phenolic content was found from the microcapsulate when the core:coating ratio was low. This probably due to the increment of the engulfing capacity of the coating material as the core concentration was reduced. Similar result was reported by (Mahdavi et al., 2016). during anthocynanin encapsulation. Therefore, the increment of the coating material concentration may improve the encapsulation efficiency of the microencapsulates.

When MD and MDHP coating materials were compared on the encapsulation efficiency, the MDHP had higher encapsulation efficiency (Fig. 1a and b). The encapsulation efficiency was ranged from 82.07–89.22% and 82.76–90.09% when MD alone and MDHP, respectively, were used as coating materials. This indicates that the mixture of coating material is more efficient than using alone. However, MD and MDHP had equivalent encapsulation efficiencies when the core coating ratio was 1:10. Contrarily, the MDHP coating material had more encapsulation efficiency when the core coating ratio was 1:8 and 1:6 (Fig. 1a). Other authors also reported that the encapsulation efficiency was improved when a mixture of coating materials were used (Sansone et al., 2011; Cilek et al., 2012; Robert et al., 2010). This might be due to the different properties of the coating materials may improve the encapsulation efficiency

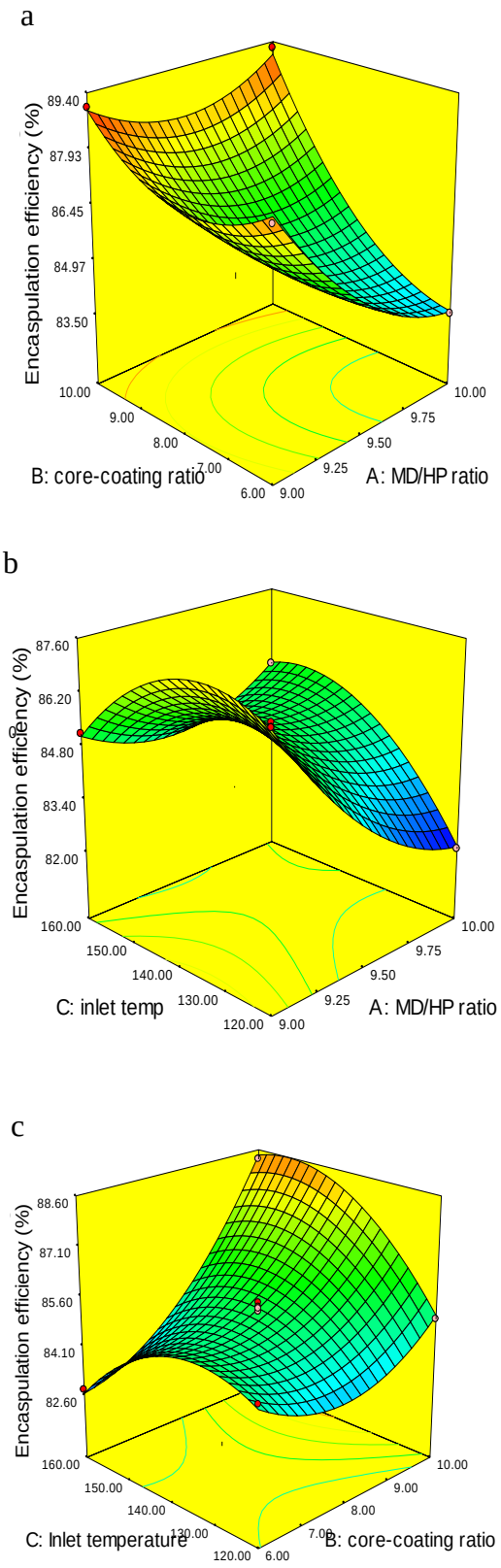

Fig.1. Response surface for the combined effects of maltodextrin:pectin (MD:HP) ratio and core:coating ratio (a); MD:HP ratio and inlet air temperature (b); core:coating ratio and inlet air temperature (c) on the encapsulation efficiency of the microencapsulates

The encapsulation efficiency was increased as an inlet air temperature was increased from 120°C to 160 °C when the core: coating ratio was 1:10 (Fig. 1c). However, when the core coating ratio was 1:6, the encapsulation efficiency was become lower. This probably due to the presence of high surface phenolic compound. Consequently, the active compounds are exposed to higher inlet air temperature. Moreover, there might be a chance of cracking on the microencapsulate surface as inlet air temperature was increased. Thus, the inner active compounds might be released and leads to the overestimation of the surface phenolic content.

$$EE = 85.28 - 1.10A + 1.58B + 0.37C + 1.15AB + 1.32AC + 1.32BC + 0.95A^2 + 1.19B^2 - 1.30C^2 \quad Eq.\ 8$$

Effects of Spray Drying Process Conditions on TPC

The TPC and TFC of the microencapsulates were increased as the core:coating ratio was increased from 1:10 to 1:6 (Fig. 2a). This is due to the increment of the concentration of the extract when the core:coating ratio was increased. Similar result was found when the core:coating ratio was changed from 1:2 to 1:1 (Tolun et al., 2016). As shown on Fig. 2a, when the core:coating ratio was 1:10, the TPC were increased, as the maltodextrin concentration was more. Conversely, as the core:coating ratio was increased to 1:8 and 1:6, the TPC was decreased. This might be due to the lowering of the encapsulation efficiency of the maltodextrin when the core:coating ratio was increased.

On the other hand, the microencapsulates coated with the mixture of maltodextrin and pectin (MDHP) had more TPC when it was compared to the maltodextrin (MD) alone. This probably due to the improvement of the encapsulation efficiency of MDHP coating material, in particular, when the core:coating ratio was increased. As described by Tolun et al. (2016), blending of maltodextrin with other coating materials improves the encapsulation efficiency and temperature sensitivity of active compounds.

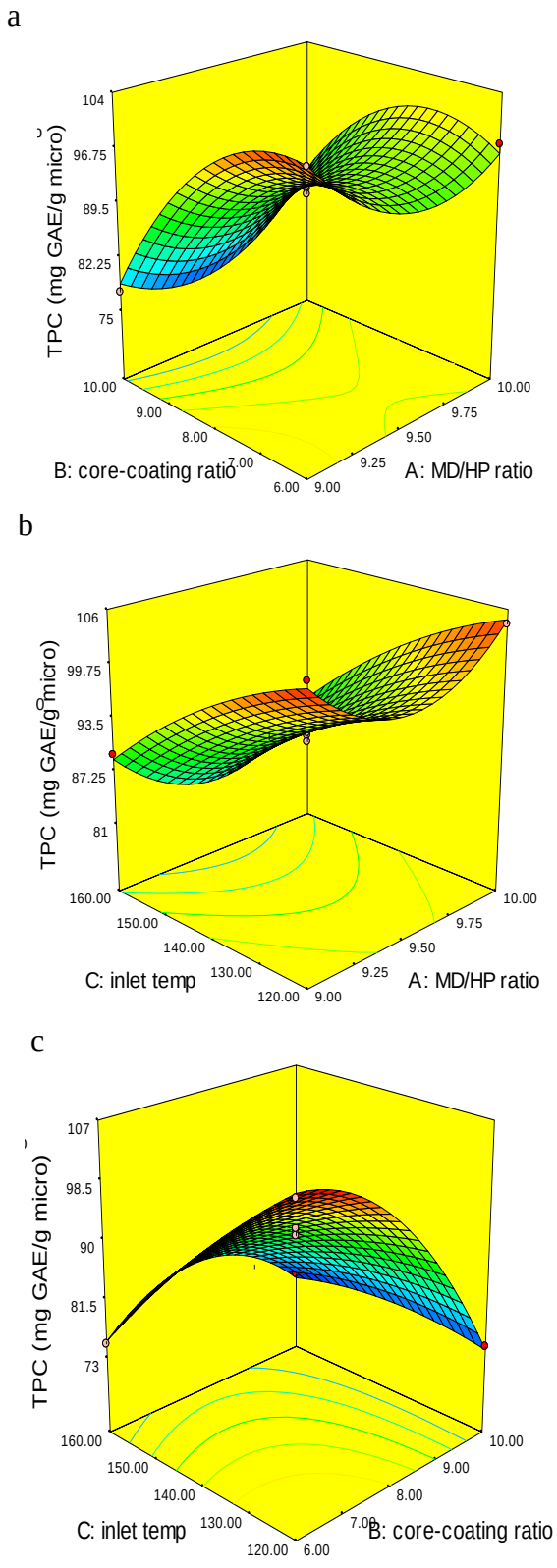

Fig. 2. Response surface for the combined effects of maltodextrin: pectin (MD:HP) ratio and core:coating ratio (a); MD: HP ratio and inlet air temperature (b); core:coating ratio and inlet air temperature (c) on the total phenolic content of the microencapsulated *M. stenopetala* product

As the shown on Fig. 2, the inlet air temperature of the spray-dryer had significant effects on the TPC of the microencapsulates. As the inlet air temperature was increased from 120 °C to 160 °C, the TPC of the microencapsulates was lowered mainly when the core:coating ratio was high. This probably due to the destruction of the active compounds at the high inlet air temperature of the spray dryer. In general, the TPC was high at an inlet air temperature of 120 °C. However, the microencapsulates developed at 140 °C had an improved encapsulation efficiency and equivalent TPC, in particular, at a core: coating ratio of 1:6 (Fig. 2). The same trend was reported by Tolun et al. (2016) in the microencapsulate from grape polyphenol. The polyphenol compounds were lost as an inlet air temperature was increased from 120 to 200 °C (Sansone et al., 2011; Tonon et al., 2008). Therefore, the bioactive compounds are sensitivity to higher inlet air temperature. As a result, the lower TPC was found when the inlet air temperature was at 160 °C. This might be due to the destruction of bioactive compounds and/or the cracking effect on the microencapsulates surface.

$$TPC = 91.89 - 8.70B - 8.07C + 3.45AB - 0.18AC + 7.60BC + 6.62A^2 - 7.59B^2 - 1.89C^2 \quad Eq.\ 9$$

Process Optimization of Spray Drying

According to the encapsulation efficiency and total phenolic content, the maltodextrin:pectin ratio (9:1), core:coating ratio (1:6), and an inlet air temperature of 140 °C were found to be the optimum spray drying process parameters for microencapsulation of bioactive compounds from *M. stenopetala* leaf extract. The predictive values with these optimum parameters are 87.8% and 103 mg GAE/g micro for encapsulation efficiency and total phenolic content, respectively.

Antioxidant Activity of the Microencapsulated Bioactive Product

The antioxidant activities of the microencapsulates were evaluated using DPPH, FRAP assays as well as ferrous chelating activity. The highest antioxidant activity was

found from the microencapsulates coated with mixture of maltodextrin and pectin than maltodextrin alone (Table 3).

The same trend was also reported by other auhors (Tolun et al., 2016; Cilek et al., 2012). Therefore, the antioxidant activities of the microencapsulates were improved when the mixture of coating materials (MDHP) were used compared to a single coating material. Regarding the core:coating ratios, the antioxidant activity was increased as the core:coating ratio was increased. This might be due to the increment of the concentration of the extract as the core:coating ratio increased. As a result, the antioxidant activities of the microencapsulates were high. On the other way, the degradation of the bioactive compounds might be higher due to the lower encapsulation efficiency. Thus, the antioxidant activity of the microencapsulated bioactive product is decreased.

The inlet air temperature of the spray dryer had a significant effect on the antioxidant activities (Table 3).

As the inlet air temperature was increased from 120 °C to 160 °C, the antioxidant activities were reduced, in particular, from the microencapsulate with the core:coating ratio of 1:6 and 1:8. This might be due to the thermal degradation of surface phenolic compounds. The result is in agreement with Tonon et al. (2008).

Moisture Content

The moisture content of the microcapsule can determine the flowability, cohesiveness as well as the storage stability of the microencapsulate (Mahdavi et al., 2016). The overall moisture content of the microencapsulate was significantly affected by coating material ratio, core:coating ratios and inlet air temperature, which was in the range of 1.38–3.56% (Table 4).

According the result, the moisture content was decreased as the inlet air temperature was increased. Other authors reported similar results (Tolun et al., 2016; Tonon et al., 2008; Laokuldilok and Kanha, 2015). This might be due the presence of the wide range of temperature gradient differences between hot air and the liquid sample, which results the higher heat transfer and higher evaporation rate. On the contrary, the increment of the inlet air temperature leads to the increment of the moisture content in Jamun juice powder (Santhalakshmy et al., 2015). This might be due to the crust formation on the drop surface' subsequently, the evaporation rate is decreased for the rest of moisture.

Table 3. Surface phenolic content and antioxidant activities of the microencapsulated bioactive product from *M. stenopetala* leaf extract

Run	Surface TPC (mg GAE/g micro)	DPPH (mg TE/g micro)	FRAP (mg TE/g micro)	Chelating (mg EE/g micro)
1	9.28±0.19 ^{g}	309.45±2.72^{fg}	260.70±1.21^{c}	41.84±1.08^{ef}
2	12.71±0.31^{de}	315.79±1.59^{e}	245.98±1.46^{e}	43.65±1.13^{ef}
3	18.70±0.64 ^{a}	383.67±2.65^{c}	281.49±1.69^{b}	54.47±1.03^{bc}
4	16.04±0.58 ^{b}	383.68±1.57^{c}	280.30±1.53^{b}	56.74±1.20^{ab}
5	8.64±0.06 ^{gh}	246.08±1.56^{j}	199.33±2.09^{g}	32.83±1.08^{g}
6	11.23±0.21^{f}	287.72±2.71^{i}	204.01±1.05^{f}	40.58±1.01^{f}
7	13.53±0.63^{c}	313.97±1.57^{ef}	254.83±2.15^{d}	50.60±1.07^{cd}
8	12.93±0.51^{cde}	306.73±2.72^{g}	242.10±1.04^{e}	42.49±1.11^{ef}
9	16.26±0.64^{b}	397.26±1.57^{b}	283.88±1.64^{ab}	57.39±0.86^{ab}
10	8.36±0.11^{h}	297.68±1.56^{h}	205.50±2.53^{f}	39.32±1.05^{f}
11	12.33±0.21^{e}	322.12±1.59^{d}	251.95±2.32^{d}	45.92±1.09^{de}
12	13.20±0.18 ^{cd}	387.30±1.39^{c}	281.89±1.34^{b}	54.17±0.99^{bc}
13	12.43±0.41^{de}	406.31±1.57 ^{a}	287.46±1.64^{a}	61.37±1.14^{a}

Table 4. Physical properties of the microencapsulated bioactive product from *M. stenopetala* leaf extract

Run	Moisture content (%)	Bulk density (g/mL)	WAI (g/g)	WSC (%)	Hygroscopicity (%)
1	3.47±0.09^{a}	0.24±0.01^{bc}	0.66±0.19^{de}	97.69±0.62^{a}	15.78±0.15^{f}
2	1.87±0.08^{ef}	0.23±0.01^{cd}	0.46±0.34^{e}	93.92±0.47^{b}	18.09±0.55^{c}
3	3.08±0.04^{b}	0.24±0.01^{abc}	0.33±0.28^{e}	93.51±0.29^{b}	15.03±0.15^{h}
4	1.76±0.03^{fg}	0.24±0.01^{bcd}	0.36±0.11^{e}	92.92±0.44^{b}	19.17±0.52^{b}
5	1.67±0.06^{gh}	0.22±0.01^{d}	1.51±0.16^{b}	85.41±0.58^{c}	15.65± 0.21^{g}
6	3.56±0.07^{a}	0.24±0.01^{abc}	1.07±0.36^{cd}	83.94±0.33^{cd}	13.43±0.52^{l}
7	2.04±0.06^{e}	0.25±0.01^{ab}	1.02±0.12^{cd}	83.45±0.59^{d}	14.83±0.27^{i}
8	1.38±0.08^{i}	0.24±0.00^{bcd}	1.34±0.18^{bc}	19.85±0.24^{de}	19.85±0.24^{a}
9	2.66±0.12^{d}	0.26±0.01^{a}	0.96±0.27^{d}	81.22±0.56^{ef}	14.86±0.51^{i}
10	2.87±0.07^{c}	0.24±0.00^{bc}	1.92±0.27^{a}	78.39±0.59^{g}	14.64±0.17^{j}
11	1.83±0.04^{fg}	0.23±0.01^{cd}	1.67±0.14^{ab}	81.25±0.83^{ef}	16.18±0.35^{e}
12	2.73±0.14^{cd}	0.24±0.00^{abc}	1.50±0.34^{b}	80.34±0.56^{f}	14.41±0.32^{k}
13	1.55±0.08^{hi}	0.25±0.01^{ab}	1.42±0.42^{b}	80.52±0.47^{f}	17.27±0.46^{d}

WAI= water absorbance index, WSC= water solubility capacity

The moisture content of the microencapsulate was increased as the core:coating ratio was changed from 1:6 to 1:10 (Table 4).

In addition, when maltodextrin and mixture of maltodextrin and pectin was used, the moisture content was found to be different (Table 4). This indicates that the moisture content of the sample depends on the type and concentration of carrier materials.

This is in agreement with the finding of Tolun et al., 2016). The decreasing of the core:coating ratio leads to the increasing of the concentration of the coating material, which in turn increases the moisture content.The higher moisture content was found when the coating material of the microcapsulate was maltodextrin than using a mixture of maltodextrin and pectin. This might be due to the relative high water binding capacity and a humectant nature of maltodextrin.

Because maltodextrin with high dextrose equivalent absorb moisture from the environment due to the presence of shorter chain polysaccharides with higher hydrophilic structure (Laokuldilok and Kanha, 2015). In general, the moisture content of the microencapsulated bioactive product was low, as a result, the storage stability of the product might be improved.

Bulk Density

The bulk density of a product is an important physical property during packaging and shipping (Fernandes et al, 2013). As shown in Table 4, the bulk density was significantly ($P < 0.05$) affected by spray drying process parameters. Lower bulk density of the microencapsulate was observed when the concentration of the coating material is higher, particularly, a mixture of coating materials was used. This might be due to higher molecular weight of the coating material (Mahdavi et al., 2016). Contrarily, the increment of the density was found when the microencapsulate was coated with maltodextrin alone. This might be related to the stickiness of property the maltodextrin. The result is in agreement with Ghosal et al. (2010), who found that the increment of maltodextrin and gum concentration in cornstarch results to lowering of the bulk density. Furthermore, Saikia et al. (2015) found a similar result when higher dextrose equivalent maltodextrin was used. On the contrary, the bulk density is decreased when the maltodextrin concentration is increased (Paini et al., 2015). This probably due to the differences in dextrose equivalent, particle size and agglomeration properties.

An inlet air temperature was considerably affect the bulk density of the microencapsulates. In general, the higher the

inlet temperature, the lower bulk density was found (Table 4). The same results were reported from encapsulated pomace polyphenol (Paini et al., 2015) and in the rice powder (Laokuldilok and Kanha, 2015). It was reasoned out that the increment of the surface area due to a higher inlet air temperature occupies a larger volume. On the contrary, Santhalakshmy et al. (2015) found that there is no significant effect of inlet air temperature on the bulk density of Jamun juice powder. The lower bulk density may indicate the exposition of the microencapsulates for oxidation due to the presence of air spaces among particles. Thus, it may result poorer storage stability of the product.

WAI

The water absorbance index (WAI) of the microencapsulated bioactive product was shown in Table 4. According to the result, the WAI was significantly increased when the coating material was mixture of maltodextrin and pectin compared to maltodextrin alone. Moreover, the WAI of the microencapsulate was increased as the core coating ratio was decreased. This might be related to the variations in the degree of engagement of hydroxyl groups to form hydrogen and covalent bonds between starch chains (Ahmed et al., 2010). Besides, it might be due to the gelling property of the pectin, which results to absorb more water. The inlet air temperature of the spray dryer also had significant effects on WAI. In general, WAI was increased as the increment of the inlet air temperature. The result was in agreement with Paini et al. (2015) who reported that higher WAI on the encapsulate of olive pomace phenols from 130 °C to 160 °C and suggested that due to the formation of amorphous structure of the coating material at high drying temperature. As cited by Ahmed et al. (2010), the increments of WAI of the product related to the loss of crystalline structure of the starch.

WSC

Water solubility capacity (WSC) of the microencapsulated bioactive product was highly affected by coating materials, core:coating ratio and inlet air temperature of the spray dryer (Table 4). The solubility of the microencapsulate coated with maltodextrin was significantly ($P < 0.05$) higher than using a mixture of maltodextrin and pectin. Furthermore, the solubility of the maltodextrin coated microencapsulate was increased as the core coating ratio was changed from 1:6 to 1:10. This indicates that the increment of the concentration of the coating material (maltodextrin) favors for the solubility of the microencapsulates. This is considerably due to the high solubility property of maltodextrin (Ahmed et al., 2010).

When an inlet air temperature was increased, the WSC of the microencapsulated product was increased (Table 4). This might be due to lower moisture content of the microencapsulate at higher inlet temperature (Tan et al., 2015), which leads to fast dissolution of the product in water. In addition, the lower inlet air temperature may result incomplete removal of moisture, thus agglomeration of the product might be formed. As reported by Goula and Adamopoulos (2008), a larger particle size is formed at a higher inlet air temperature consequently denser powder was formed that sinks in short time. Consequently, the solubility of the product is high.

Hygroscopicity

As shown on Table 4, the hygroscopicity of the microencapsulate was significantly affected by the core:coating ratio and inlet air temperature of the spray dryer. The hygroscopicity of the microencapsulate was increased as the increment of the inlet air temperature. This might be related to the moisture content of the product. It was found that at high inlet air temperature, the moisture content of microencapsulate was low. This results high moisture absorbance from the environment, thus it increases the hygroscopicity of the product. A similar result was reported by Tonon et al. (2008) from acai juice powder. When the core:coating ratio was increased, the hygroscopcity of the microencaspulate was increased (Table 4). This is probably related to

a hemactant nature of the coating material. The result is in agreement with Goula and Adamopoulos (2008) on the spray dried tomato pulp.

Color

The color values of microncapsulated bioactive product were shown in Table 5. The inlet air temperature had significant ($P < 0.05$) effect on the L^* values. As the inlet air temperature of the spray dryer was increased, the L^* values of the microencapsulates were increased. This is due to the destruction of the color pigment at higher inlet air temperature, besides, the coating material might mask the green pigment of the extract. The result is in agreement with Saikia et al. (2015) and on the spray dried Jamun juice Santhalakshmy et al. (2015). Therefore, an inlet temperature has a significant effect on the color of the powder.

The result also indicates that core:coating ratio had an effect on the L^* value. (Table 5). When the core:coating ratio was increased, the L^* value was relatively decreased. This might be due to the greenness color of the extract, which was remained on the surface the coating material. However, when the core:coating ratio was decreased, the encapsulation efficiency of the coating material was, subsequently the L^* value was increased. Cilek et al. (2012) reported the same trend with the reason that the increment of concentration of the coating material leads to suppress the color effects of the core material. In addition, when the MD and MDHP coating material is compared, the higher L^* values was shown when the MD than the MDHP coating material. This is probably due to the relative whiteness of maltodextrin and pale yellowness of the pectin. As described by Cilek et al. (2012), the whiteness of the microencapsulate was because of the white color of coating material.

Table 5. Color values of the spray dried microencapsulated bioactive product of *M. stenopetala* leaves extract

Run	L^*	a^*	b^*	Hue angle	Chroma
1	73.06±0.12^{a}	-5.19±0.03^{a}	19.03±0.05^{e}	105.25±0.64^{c}	19.73±0.20^{k}
2	71.86±0.07^{c}	-5.25±0.01^{bc}	19.29±0.11^{e}	105.21±0.94^{c}	19.99±0.17^{i}
3	67.19±0.57^{i}	-5.62±0.05^{f}	19.15±0.09^{e}	106.38±0.57^{a}	19.96±0.10^{i}
4	68.04±0.01^{h}	-5.76±0.01^{g}	19.84±0.02^{d}	106.20±0.40^{b}	20.65±0.15^{f}
5	72.37±0.09^{b}	-5.22± 0.05^{ab}	20.53±0.04^{c}	104.26±0.44^{g}	21.18±0.15^{d}
6	70.11±0.06^{f}	-5.27±0.02^{c}	20.42±0.03^{c}	104.47±0.75^{f}	21.09±0.25^{e}
7	68.75±0.07^{g}	-5.22±0.08^{ab}	20.04±0.16^{cd}	104.83±0.52^{d}	20.39±0.46^{h}
8	70.24±0.57^{f}	-5.22±0.01^{ab}	19.88±0.03^{d}	104.71±0.64^{e}	20.56±0.35^{gf}
9	68.06±0.13^{h}	-5.55±0.07^{e}	19.05±0.08^{e}	106.24±0.20^{b}	19.84±0.17^{j}
10	71.15±0.57^{d}	-5.22±0.04^{ab}	21.67±0.07^{a}	103.53±0.82^{i}	22.29±0.31^{a}
11	71.12±0.08^{d}	-5.22±0.08^{b}	20.46±0.15^{c}	104.32±0.52^{g}	21.12±0.10^{de}
12	70.66±0.06^{e}	-5.24±0.05^{bc}	21.24±0.08^{ab}	103.86±0.32^{h}	21.88±0.12^{b}
13	70.16±0.01^{f}	-5.45±0.01^{d}	21.11±0.02^{b}	104.48±0.94^{f}	21.80±0.20^{c}

The color value of a^* indicated that the inlet air temperature had a significant effect (Table 5). The a^* value was shown with a negative sign that indicates the value placed in the second quadrant on the color coordinates ($-a^*$, b^*). The small value of a^* indicates the lowering of the greenness color. The increment of the inlet air temperature leads to have the small a^* value. This is due to the destruction of the green pigment at higher inlet air temperature. Whereas, the value was high when the core coating ratio was decreased. This might be due to the higher encapsulation efficiency of the microcapsulate which in turn it masks the green color of the extract. On the other hand, the b^* values of the spray dried microencapsulate coated with a mixture of maltodextrin and pectin was significantly higher when it was

compared to the maltodextrin alone. This is due to the slight yellowness of the pectin material. In addition, when the core:coating ratio was decreased, the *b** value was higher. This is due to the increment of the coating material. However, the inlet temperature had slight effects on the *b** values. The result is agreed with Santhalakshmy et al. (2015) who found that *b** values were changed insignificantly reported a similar result.

The color of the product is determined using the hue angle, whereas the color intensity is by using the Chroma value (Deore et al., 2013). The coating materials, core:coating ratios and inlet air temperature significantly affect the Hue angle and Chroma values of the spray-dried microcapsules. The lower hue angle was observed when the inlet air temperature increased and the opposite is true for Chroma values (Table 4). This is probably because of the degradation of the pigment compounds at a higher inlet air temperature as well as higher encapsulation efficiency of the extract. This is in agreement with Santhalakshmy et al. (2015) during encapsulation of the anthocyanin compounds.

4. CONCLUSIONS

The optimal spray drying process conditions were predicted to encapsulate the bioactive compounds from *M. stenopetala* leaf extract. The best spray drying process conditions were maltodextrin: pectin ratio 9:1 (w/w), core: coating ratio 1:6 (w/w) with an inlet air temperature of 140 °C. The predicted values with these parameters are 87.8% for encapsulation efficiency with the TPC values of 103.55 mg GAE/g micro. This study showed that the spray drying process conditions considerably affect the physical and functional properties and the bioactive contents of the microencapsulated bioactive product from *M. stenopetala* leaf extract.

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