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Optimization of Spray for Drying *Morinda citrifolia* L. Fruit Extract

D. Krishnaiah, R. Sarbatly and R. Nithyanandam
Phytochemical Laboratory, Department of Chemical Engineering,
School of Engineering and Information Technology, Universiti Malaysia Sabah, 88999,
Kota Kinabalu, Malaysia

Abstract: Hot water extract of *Morinda citrifolia* L. fruit was spray dried using κ-carrageenan (1 wt%). Spray drying was carried out according to D-optimal design and independent variables selected were temperature and M_{core}/M_{wall}. Spray drying process was optimized by using Response Surface Methodology (RSM) for four different responses such as moisture content, DPPH scavenging activity, Total Phenolic Content (TPC) and Total Flavonoid (TF). Effects of temperature and ratio of core to wall material found to be significant on all responses. Applying desirability function method, optimal spray drying condition for κ-carrageenan as binding material were found to be 1:1.5 (M_{core}/M_{wall}) at 90°C. Experimental value of response variables match well with the predicted values. The nanoparticles obtained in this study represent an interesting food additive for incorporation into functional foods due to presence of antioxidants.

Key words: *Morinda citrifolia*, microencapsulation, spray-drying, antioxidant, response surface methodology

INTRODUCTION

Morinda citrifolia L. (Noni) has been used in traditional Polynesian medicine for more than 2000 years. This plant belongs to the family Rubiaceae and is commonly found in the Hawaiian and Tahitian islands. The bark, stem, root, leaf and fruit have been used traditionally as folk remedies for many diseases, including diabetes, hypertension and cancer (Hirazumi *et al*., 1994, 1996). These diseases are caused by the generation of Reactive Oxygen Species (ROS). This ROS generation normally occurs when the body is under stress. In traditional pharmacopoeia, *M. citrifolia* L. fruit is claimed to prevent and cure several diseases. It is primarily used to stimulate the immune system and thus to fight bacterial, viral, parasitic and fungal infections. It is also used to prevent the formation and proliferation of tumours, including malignant ones (Dixon *et al*., 1999).

Levand and Larson (1979) identified several compounds including acetyl derivatives of asperuloside, glucose, caproic acid and caprylic acid in *M. citrifolia* L. fruits. Wang *et al*. (1999) isolated two glycosides, rutin and asperulosidic acid and a novel trisaccharide fatty acid ester from *M. citrifolia* L. fruits. Carboxylic acids, especially octanic acid and hexaonic acid, have also been identified from the fruits. Based on the literature, it is clear that the fruit has antibiotic and antioxidant properties, but there is a lack of scientific evidence based on *in vivo* studies. Because *M. citrifolia* L. is believed to have high antioxidant potential, it can substitute as an antioxidant compound in food items and in preventive medicines. In our recent work, the antioxidant potentials of various medicinal plant species were reviewed (Krishnaiah *et al*., 2010).

The microencapsulation technique of spray-drying is an effective way to protect drug or food ingredients against deterioration and volatile losses. The protective mechanism consists of the formation of a membrane wall that encloses droplets or particles of the encapsulated material. There are currently a variety of means available to prepare such microparticles. The choice of a particular method depends on the type of microparticles desired. The properties of the wall structures, size and shape are important considerations. However, in the food and drug industry, spray-drying is still the most popular method of forming microparticles because it is very easy to industrialise and allows for continuous production (Su *et al*., 2008). Hence many researchers have used spray drying technique for the production of microparticles of fruit extract (Tonon *et al*., 2010; Saenz *et al*., 2009; Kha *et al*., 2010; Goula and Adamopoulos, 2010; Krishnaiah *et al*., 2009). Neverthless, micro-encapsulation of *M. citrifolia* L. extract by spray-drying has not been reported to date.

For microencapsulation by spray-drying, carrageenan is a good choice as the wall material due to its pseudoplastic properties. These properties allow it to act as a plasticiser, promoting the formation of spherical and

Corresponding Author: D. Krishnaiah, Phytochemical Laboratory, Department of Chemical Engineering, School of Engineering and Information Technology, Universiti Malaysia Sabah, 88999, Kota Kinabalu, Malaysia Tel: +60-143749743 Fax: +60-88-320348

smooth-surfaced microcapsules and enhancing the adhesion force between the wall and the core materials (Su *et al*., 2008). Moreover, it has the desirable properties of emulsification, edibility and biodegradation. Carrageenan is a polysaccharide, which is often used as an excipient in the drug industry.

The Response Surface Methodology (RSM) has been demonstrated to be a powerful tool for determining the factors effects and their interactions, which allow process optimization to be conducted effectively (Bas and Boyaci, 2007). This method is the preferred experimental design for fitting polynomial model to analysis the response surface of multi-factor combinations ie., RSM uses quantitative data from appropriate experiments to determine and simultaneously solve multivarient equations (Myers *et al*., 2004). Generally speaking, RSM attempts to fit a polynomial of appropriate to the response of the system of interest. The goal of the system of interest is termed the response. This response is normally measured on a continuous scale and is a variable which likely represents the most important function of the system, though this does not rule out the possibility of investigating more than one system function, i.e., more than one response. Also contained in the system are input variables that affect the response and are subject to control (Vashi *et al*., 1995).

The response surface procedures involve experimental strategy, mathematical methods and statistical inference which, when combined, enable users to make an efficient empirical exploration of the system in which they are interested (Meyers, 1976). The experimental strategy enables the analyst to explore the response surface with equal precision, in any direction. The experimental design initially limits the region under investigation. Subsequent to the initial investigation, the experimental design enables the analyst to explore the response surface in a systematic manner in the direction that offers the most promise for improvement (Awang *et al*., 2004). RSM offers techniques for finding the optimum response of the system in an efficient manner (Powers, 1989). The major advantage of RSM is the amount of data needed for evaluation, analysis and optimization. It significantly reduces the number of experiments required. RSM is a faster and more economical method for gathering research results than classic one-variable at a time or full-factors experimentation. The RSM software, design expert 8.0.2 has been used for this purpose. RSM generated the experimental design table using D-optimal method.

The objective of the present study was to elucidate the effect of adjuvant in the spray-drying of noni fruit extract and to find the optimal processing parameters of spray-drying to create powdered noni extract with the best free radical scavenging activity, the highest total phenolic content, the highest total flavonoid content as well as the best particle size and moisture content by applying RSM.

MATERIALS AND METHODS

Chemicals: Methanol and ethyl acetate (HPLC grade) were obtained from J.T. Baker (USA). Folin Ciocalteau reagent and tannic acid (TA) were purchased from Merck, Germany. The chemicals 2, 2-diphenyl picryl hydrazyl (DPPH), aluminiumtrichloride ($AlCl_3$) and sodium carbonate were obtained from Sigma Chemicals (St. Louis, USA). A total of 5 kg of *M. Citrifolia* L. fruit before the ripening stage was obtained from Kota Kinabalu, Sabah, Malaysia. All other chemicals were of reagent grade and were used without further purification.

Preparation of fruit extracts: *M. citrifolia* L. fruit was washed with tap water followed by washing with distilled water. The fruit was peeled and the core (pulp and seed) was cut into small species. The skin, pulp and seed were sun-dried for two days. Then, the sample was kept at 60°C in a hot-air oven for one day to remove all moisture. The dried fruit was then finely powdered using a mixer. A total of 250 mL of ethyl acetate was added to 25 g of powdered sample (10 wt%) and extraction was performed in a water shaker bath at 35°C for three days. The supernatant was then separated from the residue by filtration using Whatman No. 1 filter paper. The extracted solution was stored in a closed container and kept at 4°C before being analysed.

Spray-drying of the noni fruit extract: κ-Carrageenan (1 wt.%) were mixed with *M. citrifolia* L. fruit extract at different volume ratios (1:1, 1:1.6 and 1:4 for κ-carrageenan) as per experimental design conditions of RSM and then stirred to form an aqueous solution. The resulting emulsion was then spray-dried using a lab plant spray-dryer SD-05 (pilot scale) with co-current flow (the spray-dried product and the drying air flow were in the same direction). The drying chamber had a diameter of 215 mm and a height of 500 mm. The main components of the system were the feed system of the *M. citrifolia* L. fruit extract consisting of a peristaltic pump, a fluid atomiser (inlet orifice diameter of 0.5 mm) and an air compressor as well as a feed system for drying the gas consisting of a blower and an air filter. Finally, a temperature control system and a product control system (cyclone) were also employed. The feed flow rate was kept at 315 mL h^{-1}. The flow rate of the drying air was fixed at

60 $m^3 h^{-1}$ and the atomising air remained at a pressure of 1.1 bar. After spray-drying, the powders were collected through a high efficiency cyclone in a glass container, transferred to a glass vial and stored in desiccators at ambient temperature.

Several spray-drying runs were carried out as per D-optimal experimental design to investigate the effects of the inlet drying gas temperature for different volume ratios of *M. citrifolia* L. fruit extract to excipient. Further analyses were performed to determine the antioxidant activity, the total phenolic content and the total flavonoid content of the spray-dried powder.

Moisture content: The moisture content of the spray-dried microparticles was determined by the oven drying method. Samples of the microparticles with predetermined masses were placed in an oven (Memmert), heated to 102°C and weighed on an analytical balance (Mettler Toledo PB153-S/FACT, Switzerland) until a constant mass was observed. The product moisture content was determined from the weight loss by averaging three measurements.

DPPH radical scavenging activity: DPPH is a stable free radical that reacts with compounds that are able to donate a hydrogen atom. Thus, the hydrogen donating abilities of spray-dried *M. citrifolia* L. fruit extract was determined from the change in absorbance at 515 nm by the Blois (1958) method with slight modifications. For free radical scavenging measurements, samples in methanol solution were prepared by dissolving 10 mg of spray-dried powder in 30 mL of methanol and centrifuging for 10 min using a Sartorius Sigma 3-18 K centrifuge. Aliquots of supernatant were added to 3 mL of 0.025 g L^{-1} DPPH in methanol. The change in absorbance was measured after 40 min at room temperature using a 4802 UV-VIS double-beam spectrophotometer. Methanol was used as the reference. DPPH (0-100 mg L^{-1}) was used to obtain a standard calibration curve. All measurements were made in triplicate. The DPPH radical scavenging activity in terms of a percentage was calculated according to the following equation:

$$\text{DPPH scavenging activity (\%)} = 1 - \frac{\text{Abs}_{515}\ \text{sample}}{\text{Abs}_{515}\ \text{DPPH solution}} \times 100\%$$

Total phenolic content: The total phenolic content (TPC) was determined according to the Folin Ciocalteau method (Slinkard and Singleton, 1977) with slight modifications. Briefly, samples in methanol solutions were prepared by dissolving 10 mg of spray-dried powder in 30 mL of methanol and centrifuging for 10 min using a Sartorius Sigma 3-18 K centrifuge. The supernatant of the sample extract (0.5 mL) was added to 2.5 mL of the 0.2 N FC reagent and allowed to react for 5 min. Then, 2 mL of 75 g L^{-1} sodium carbonate was added to the reaction mixture and diluted to 25 mL using distilled water. Finally, the reaction mixture was incubated for two hours at room temperature and the absorbance was measured at 760 nm using a 4802 UV-VIS double-beam spectrophotometer. Methanol was used as the reference. Tannic acid (0-100 mg L^{-1}) was used to produce a standard calibration curve. The total phenolic content was expressed in mg of tannic acid equivalents (TAE/g of spray-dried powder).

Total flavonoid content: The total flavonoid content was determined using the Dowd method as adopted by (Arvouet-Grand *et al*., 1994). A total of 5 mL of 2% aluminium trichloride ($AlCl_3$) in methanol was mixed with the same volume of the extract solution (0.4 mg mL^{-1}). Absorption readings at 415 nm using a UV-VIS double-beam spectrophotometer were taken after 10 min against a blank sample consisting of a 5ml extract solution with 5 mL of methanol without $AlCl_3$. The total flavonoid content was determined using a standard curve with catechin (0-100 mg L^{-1}) as the standard. The content is expressed as mg of catechin equivalents (CE g^{-1} of extract).

Experimental design: Response surface methodology (RSM) was applied using a commercial statistical package, Design expert version 8.0.2 (Statease Inc., Minneapolis, USA) to identify optimum levels of two variables of temperature (°C) and M_{core}/M_{wall} (no unit) regarding of four responses-moisture content, DPPH scavenging activity, TPC and TF of spray dried *M. citrifolia* L. fruit extract. The coded and uncoded independent variables used in the RSM design are listed in Table 1. The experiments were designed according to the D-optimal design as shown in Table 2. The order of experiments has been fully randomized. Data were analyzed by multiple regressions through the least-square method.

A cubic model was used to express the responses as a function of independent variables, where A and B are coded values of temperature and M_{core}/M_{wall}. The test of statistical significance was used on the total error

Table 1: Factors and levels tested for the experimental design

Parameter	Temperature (°C)	M_{core}/M_{wall}
Factor	A	B
Max. parameter	140	1
High level	+1	
Average parameter	115	0.625
Medium level	0	
Min. parameter	90	0.25
Low level	-1	

Adjuvant/Binding material: κ-carrageenan

criteria with a confidence level of 95%. The significant terms in the model were found by analysis of variance (ANOVA) for each response. The adequacy of model was checked accounting for R^2 and adjusted-R^2. Numerical and graphical optimization technique of the design expert software was used for simultaneous optimization of the multiple responses. The desired goals for each variables and response were chosen. All the independents variables were kept within range while the responses were either maximized or minimized.

RESULTS AND DISCUSSION

Model fitting: The responses of MC, DPPH scavenging activity, TPC and TF obtained from all the experiments are listed in Table 2. The experimental data were used to calculate the co-efficients of cubic equation and Table 3 and 4 summarizes the results of ANOVA-the significance of the coefficients of the models and regression co-efficient respectively. For any of the terms in the model, a large regression co-efficient and a small p-value would indicate a more significant effect on the respective response variables. ANOVA showed that the resultant cubical model adequately represented the experimental data with the coefficient of multiple determination (R^2) of 0.89, 0.98, 0.65, 0.58 for the response of MC, DPPH activity, TPC and TF.

Coefficient of determination (R^2) is the proportion of variation in the response attributed to the model rather than to random error and was suggested that for a good fitted model, R^2 should not be less than 80%. When R^2 approaches to the unity, signifies the suitability of fitting empirical model to the actual data. The lower value of R^2 shows the inappropriateness of the model to explain the relation between variables (Little and Hills, 1978; Mendenhall, 1975). It should be noted that adding a variable to the model will always increase R^2, regardless of whether the additional variable is statistically significant or not. Thus, a large value of R^2 does not always imply the adequacy of the model. For this reason, it is more appropriate to use an adj-R^2 of over 90% to evaluate the model adequacy. Only for DPPH scavenging activity the adj- R^2 values were found to be higher than 0.90. Higher adj- R^2 indicated that non significant terms have not been included in the model. Each of the experimental value is compared to the predicted value calculated from the model (Table 2). The results suggest that the models used in this research were able to identify the optimum operating condition of spray drying of *M. citrifolia* L. fruit extract.

Response surface analysis of moisture content: Response Surface Analysis (RSA) of the data in Table 2 demonstrates that the relationship between the moisture content and independent variables is cubic with good regression coefficient (R^2 = 0.89). Equation 1 shows the relationship between moisture content and independent variables (temperature and M_{core}/M_{wall}):

$$MC = 6.61-1.20A-0.15B + 0.08AB + 1.48A^2 + 1.63B^2- 1.59A^2B + 3.59AB^2 \quad (1)$$

Figure 1(A) is a response surface plot showing the effect of temperature and M_{core}/M_{wall} on the moisture content. Both independent variables showed a positive cubic effect (AB^2) on the moisture content ($p<0.01$). The moisture content first increased and then decreased when M_{core}/M_{wall} increased, which revealed that higher ratio is favorable to obtain low moisture content at lower temperature. At higher temperature, the moisture content first decreased and then increased when M_{core}/M_{wall} increased, which revealed that ratio of 0.55 is favorable to obtain low moisture content. As the temperature increases at the lower ratio of M_{core}/M_{wall} (more binding material), the moisture content increase steeply this may due to high hygroscopicity of the powder obtained. However, in the experimental region the lowest moisture content was observed in 1:1 at 90°C.

Response surface analysis of DPPH activity: The RSA in Table 2 demonstrates that the relationship between the DPPH activity and independent variables is cubic with good regression coefficient (R^2 = 0.98). Equation 2 shows the relationship between DPPH activity and independent variables (temperature and M_{core}/M_{wall}):

$$DPPH = 10.77-8.48A+0.43B+6.60AB+6.19A^2- 2.56B^2-1.57A^2B+9.18AB^2 \quad (2)$$

Figure 1(B) is a response surface plot showing the effect of temperature and M_{core}/M_{wall} on the DPPH scavenging activity. Temperature showed a negative linear effect and positive quadratic effect on the DPPH scavenging activity ($p<0.0001$;$p<0.001$). M_{core}/M_{wall} showed a negative quadratic effect on the DPPH scavenging activity ($p<0.05$). Both independent variables showed a positive interaction effect and positive cubic effect (AB^2) on the above response ($p<0.0001$; $p<0.0001$). The DPPH scavenging activity first increased and then decreased when M_{core}/M_{wall} increased, which revealed that medium ratio is favorable to obtain high DPPH scavenging activity at lower temperature. At higher temperature, the DPPH scavenging activity increased steeply when M_{core}/M_{wall} increased, which revealed that ratio of 1 is favorable to obtain high DPPH scavenging activity. As the temperature increases at the lower ratio of M_{core}/M_{wall} (more binding material), the DPPH scavenging

Table 2: Experimental design conditions and results

	Two factors											
	Factor 1	Factor 2			Responsive value							
	Temperature (°C)	M_{core}/M_{wall} (no unit)	Factor 1 A	Factor 2 B	MC%		DPPH scavenging activity (%)		TPC mg of TAE/ g of SDP		TF mg of CE/ g of SDP	
S. No.	Coded values		Coded values		AV	PV	AV	PV	AV	PV	AV	PV
1	140.00	0.625	+1	0	5.50	6.89	7.64	8.48	21.00	19.03	17.50	18.22
2	90.00	0.625	-1	0	8.70	9.29	24.91	25.44	30.00	27.51	25.00	23.36
3	140.00	0.250	+1	-1	15.00	13.77	10.24	9.64	21.00	23.33	22.50	23.88
4	90.00	0.250	-1	-1	9.50	9.15	21.66	21.44	21.00	22.99	17.50	22.08
5	140.00	0.250	+1	-1	14.00	13.77	9.50	9.64	24.00	23.33	22.50	23.88
6	90.00	1.000	-1	+1	5.90	5.51	5.85	5.96	27.00	25.99	30.00	26.08
7	90.00	0.250	-1	-1	9.50	9.15	21.66	21.44	24.00	22.99	27.00	22.08
8	90.00	0.625	-1	0	8.50	9.29	25.12	25.44	27.00	27.51	21.00	23.36
9	115.00	1.000	0	+1	6.70	8.09	7.80	8.64	27.00	25.03	30.00	30.72
10	90.00	1.000	-1	+1	5.80	5.51	6.50	5.96	24.00	25.99	22.50	26.08
11	115.00	0.250	0	-1	7.00	8.39	6.94	7.78	21.00	19.03	30.00	30.72
12	140.00	1.000	+1	+1	11.40	10.45	20.07	20.56	21.00	23.01	27.00	26.84
13	140.00	1.000	+1	+1	10.20	10.45	21.50	20.56	24.00	23.01	27.00	26.84
14	140.00	0.250	+1	-1	13.00	13.77	9.60	9.64	24.00	23.33	27.00	23.88
15	115.00	0.625	0	0	7.50	6.61	9.24	10.77	21.00	21.47	25.00	26.79
16	115.00	0.625	0	0	8.50	6.61	14.00	10.77	18.00	21.47	30.00	26.79

AV: Actual values; PV: Predicted values

Table 3: Variance analysis for responses

Sov	Sum of square	Degrees of freedom	Mean square	F-value	p-value Prob>F
MC					
Regression	113.74	7	16.25	8.82	0.0032
Residual	14.74	8	1.84		
Total	128.47	15			
DPPH scavenging activity					
Regression	763.57	7	109.08	50.80	<0.0001
Residual	17.18	8	2.15		
Total	780.74	15			
TPC					
Regression	96.27	7	13.75	2.13	0.1557
Residual	51.67	8	6.46		
Total	147.94	15			
TF					
Regression	152.83	7	21.83	1.58	0.2661
Residual	110.28	8	13.79		
Total	263.11	15			

activity decreases steeply and increases slightly further. From the results it was clear that the activity is higher either medium ratio at lower temperature or higher ratio at higher temperature, this may due to at these points the ingredients binding effectively with wall material. However, in the experimental region the highest DPPH scavenging activity was observed in 1:1 at 140°C.

Response surface analysis of total phenolic content: The RSA of the data in Table 2 demonstrated a regression value (R^2 = 0.65) and Eq. (3) showed the relationship between TPC and independent variables (temperature and M_{core}/M_{wall}):

$$TPC = 21.47-4.24A+3.00B-0.83AB+1.80A^2+ 0.56B^2-2.33A^2B + 3.58AB^2 \quad (3)$$

Figure 1(C) is a response surface plot showing the effect of temperature and M_{core}/M_{wall} on the total phenolic content. Temperature showed a negative linear effect on the total phenolic content ($p<0.05$). The TPC increased gradually when M_{core}/M_{wall} increased, which revealed that higher ratio is favorable to obtain high total phenolic content at lower temperature. At higher temperature, the total phenolic content first decreased and then increased to the same level when M_{core}/M_{wall} increased, which revealed that ratio of either 0.25 or 1 is favorable to obtain high phenolic content. As the temperature increases at the lower ratio of M_{core}/M_{wall} (more binding material), TPC decreases and then increases, attain a maximum value at 140°C. From the results it was clear that as that of DPPH activity, higher ratio at higher temperature, TPC is also maximum this may due to at these points the ingredients

Table 4: Estimated co-efficients for the experimental design and their VIF

Co-efficient	Value	Standard error	Confidence interval at 95% confidence		VIF
			Low	High	
MC					
Intercept	6.61	0.78	4.81	8.42	
A-Temperature	-1.20	0.82	-3.09	0.69	4.37
B-Mcore/Mwall	-0.15	0.96	-2.36	2.06	5.47
AB	0.08	0.46	-0.98	1.14	1.02
A^2	1.48	0.82	-0.40	3.36	1.08
B^2	1.63	0.78	-0.17	3.42	1.13
A^2B	-1.59	1.06	-4.04	0.86	5.49
AB^2	3.59	0.94	1.42	5.76	4.30
R^2	0.89				
Adj-R^2	0.78				
DPPH scavenging activity					
Intercept	10.77	0.84	8.83	12.72	
A-Temperature	-8.48	0.88	-10.52	-6.44	4.37
B-Mcore/Mwall	0.43	1.04	-1.96	2.82	5.47
AB	6.60	0.50	5.46	7.75	1.02
A^2	6.19	0.88	4.16	8.22	1.08
B^2	-2.56	0.84	-4.49	-0.62	1.13
A^2B	-1.57	1.15	-4.22	1.08	5.49
AB^2	9.18	1.02	6.83	11.52	4.30
R^2	0.98				
Adj-R^2	0.96				
TPC					
Intercept	21.47	1.46	18.09	24.85	
A-Temperature	-4.24	1.53	-7.78	-0.70	4.37
B-Mcore/Mwall	3.00	1.80	-1.14	7.14	5.47
AB	-0.83	0.86	-2.82	1.15	1.02
A^2	1.80	1.53	-1.72	5.32	1.08
B^2	0.56	1.46	-2.80	3.92	1.13
A^2B	-2.33	1.99	-6.93	2.26	5.49
AB^2	3.58	1.76	-0.49	7.64	4.30
R^2	0.65				
Adj-R^2	0.35				
TF					
Intercept	26.79	2.14	21.85	31.72	
A-Temperature	-2.57	2.24	-7.74	2.60	4.37
B-Mcore/Mwall	0.00	2.63	-6.05	6.05	5.47
AB	-0.26	1.26	-3.16	2.63	1.02
A^2	-6.00	2.23	-11.15	-0.86	1.08
B^2	3.93	2.13	-0.98	8.84	1.13
A^2B	1.74	2.91	-4.98	8.45	5.49
AB^2	3.21	2.57	-2.73	9.15	4.30
R^2	0.58				
Adj-R^2	0.21				

binding effectively with wall material. However, in the experimental region the highest total phenolic content was observed in 1:1 at 90°C.

Response surface analysis of total flavonoid: The RSA in Table 2 demonstrated a low regression value (R^2 = 0.58) and Eq. (4) showed the relationship between TF and independent variables (temperature and M_{core}/M_{wall}).

$$TF=26.79-2.57A-0.26AB-6.00A^2+3.93B^2+1.74A^2B+3.21AB^2 \quad (4)$$

Figure 1(D) is a response surface plot showing the effect of temperature and M_{core}/M_{wall} on the total flavonoid. Temperature showed a negative quadratic effect on the total flavonoid ($p<0.05$). The TF increased gradually when M_{core}/M_{wall} increased, which revealed that higher ratio is favorable to obtain high total flavonoid at lower temperature. At higher temperature, the total flavonoid first decreased and then increased when M_{core}/M_{wall} increased, which revealed that ratio of 1 is favorable to obtain high flavonoid content. As the temperature increases at the lower ratio of M_{core}/M_{wall} (more binding material), TF increases and then decreases, attain a maximum value at 120°C. From the results it was clear that higher ratio at medium temperature, TF is maximum. However, in the experimental region the highest total flavonoid was observed in 1:1 at 120°C.

Optimization of spray drying operating condition: The spray drying condition would be optimum if the DPPH scavenging activity, TPC and TF reached maximum values

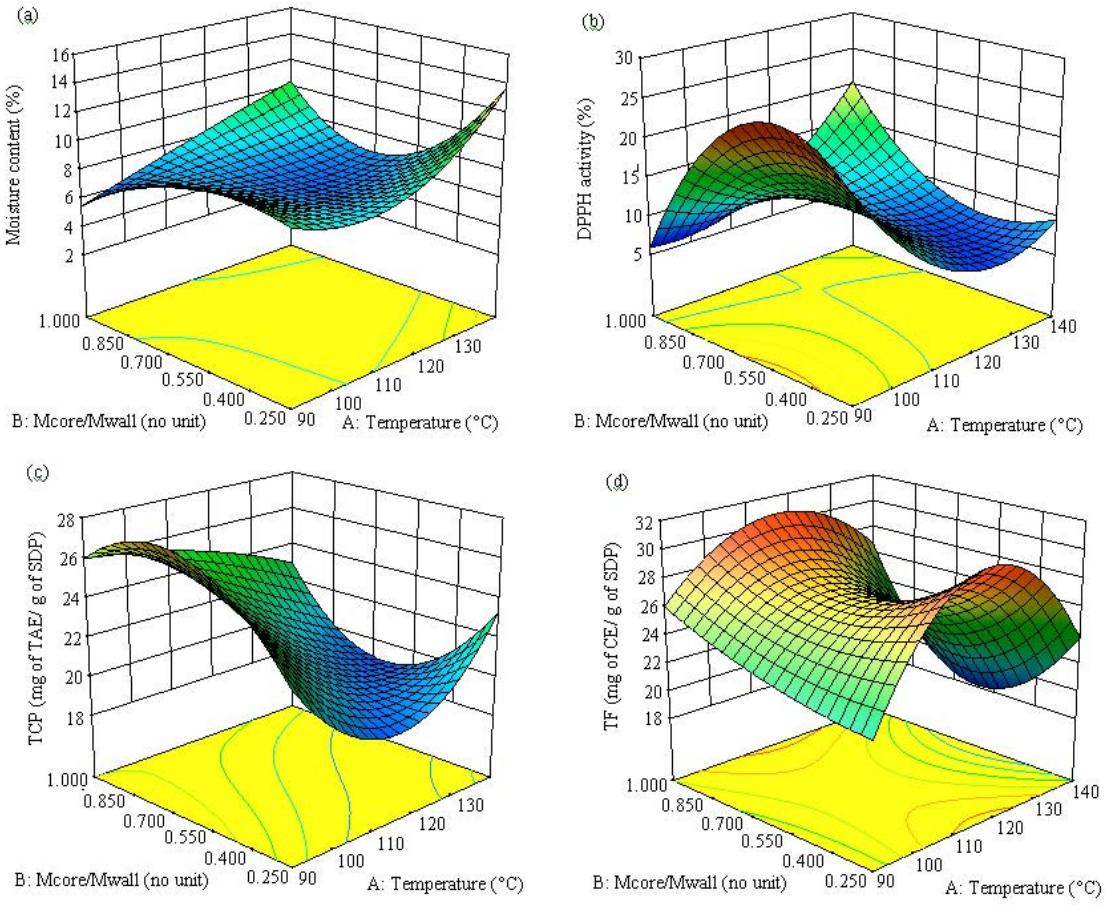

Fig. 1: Output experimental design variations with temperature and M_{core}/M_{wall}, (κ-carrageenan, (a) moisture content, (b) DPPH scavenging activity, (c) TPC and (d) TF)

and moisture content reached minimum values. The values of all the responses at operating condition were converted to a desirability function. The desirability values of the minimum and maximum were configured as 0 and 1 respectively. Maximum desirability function obtained was taken as optimum operating condition. Optimal spray drying condition for κ-carrageenan as binding material were found to be 1:1.5 (M_{core}/M_{wall}) at 90°C.

CONCLUSION

With the advent of statistical techniques such as RSM, especially as software packages for problem-solving such as optimization of the processes, products, design etc., have reduced the time required to perform those operations not only by the speed of the computer but also by reducing the quantity of experimental data required for optimization analysis. They have lead to better understanding of the process through results represented in the form of graphs such as 3D surface, contour etc. Many experiments have been conducted on spray drying of *M. citrifolia* L. fruit extract for different operating conditions: temperature (90-140°C) and M_{core}/M_{wall} (1:1 to 1:4). An experimental design has been constructed as per RSM D-optimal design conditions in order to study the influence of above parameters on moisture content, DPPH scavenging activity, TPC and TF. RSM and the conventional graphic and desirability functions methods have been effective in determining the optimum zone within the experimental region. From the response surface cubic model it was found that spray drying condition was significantly affected by temperature and M_{core}/M_{wall}. At optimum spray drying condition of κ-carrageenan, moisture content, DPPH scavenging activity, total phenolic content and total flavonoid were found to be 9.13, 24.71%, 27.62 mg of TAE g^{-1} of SDP and 23.53 mg of CE g^{-1} of SDP

respectively. This study also reveals that *M. citrifolia* L. fruit extract is a good source of antioxidant, phenolic compounds and flavonoids.

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