Original Article
Food Science and Technology

ISSN 0101-2061 (Print)
ISSN 1678-457X (Online)

DOI: https://doi.org/10.1590/fst.102621

Production of instant pomelo peel powder by spray drying: Optimization of wall material composition to microencapsulate phenolic compounds

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Abstract

Microencapsulation technique helps to protect the core materials from deterioration, thereby improving the bioavailability of active compounds. In microencapsulation, the wall properties determine the encapsulation efficiency, and better results can be achieved when a mixture of multiple wall materials is used. This study aimed to optimize the wall material composition of pomelo peel (*Citrus maxima* (Burn.) Merr.) microcapsules prepared by spray drying technology to achieve the best values of polyphenol recovery and encapsulation efficiency. Response surface methodology was employed and a Box-Behnken design was used to investigate the effects of four independent variables, including the concentrations of resistant maltodextrin, pectin, β-cyclodextrin, and gum arabic. The concentration of the feeding liquid, and spray drying inlet and outlet temperatures were fixed at 30 °Bx, 180 °C, and 65 °C, respectively. The results showed that the optimized wall material composition consisted of 52.66% maltodextrin, 0.92% pectin, 5.30% β-cyclodextrin, and 6.28% gum arabic. Using this formula, the polyphenol recovery yield was found to be 78.86%, while the encapsulation efficiency was 77.78%, which agreed well with the predicted values of 78.90% and 77.67%. These results strongly indicate that the optimized wall material composition proposed in this study may be beneficial in the encapsulation process.

Keywords: Box-Behnken design; *Citrus maxima* (Burn.) Merr.; microencapsulation; polyphenol; response surface methodology; wall material composition.

Practical Application: Microcapsules can be used as a source of natural pigments or as nutraceutical products.

1 Introduction

Pomelo (*Citrus maxima* [Burn.] Merr.) is an economically significant citrus fruit tree that is widely cultivated in the Mekong Delta of Vietnam (Nguyen et al., 2021b; Tran et al., 2020). However, its peel is also a primary source of agricultural waste, as the peel can weigh up to 48% of the whole fruit (Joglekar et al., 2019; Tran et al., 2020). Due to fermentation, this waste causes many economic and environmental problems (Fayek et al., 2017; Joglekar et al., 2019). Therefore, instead of composting, recovering phenolic compounds is an alternative way to make use of this citrus waste to create value-added products (Londoño-Londoño et al., 2010; Nguyen et al., 2021a; To & Nguyen, 2021). Several phenolic compounds, such as flavanones (hesperetin and naringenin), flavone glycosides (hesperidin and naringin), and polymethoxylated flavones (PMF), have been discovered in citrus peels (Londoño-Londoño et al., 2010; Manthey, J. A., & Guthrie, 2002). They may be used as antiproliferative, antidiabetic, and hypocholesterolemic nutraceutical products (Fayek et al., 2017), or/and as anti-hyperglycemic pharmaceutical alternatives (Tran et al., 2021).

However, phenolic compounds are sensitive and can be easily affected by many physicochemical factors (Fang & Bhandari, 2010; Xu et al., 2019), which creates a significant challenge for their incorporation into food products (Fang & Bhandari, 2010; Vo et al., 2019). The microencapsulation of phenolic compounds is an effective strategy to overcome this problem. By coating the sensitive compounds within the carrier material, this technique can protect the core from deterioration, thereby improving the bioavailability of active compounds (Baranauskaite et al., 2019; Garcia et al., 2020; Pai et al., 2015; Santos et al., 2020b). This technique can also help to mask the unpleasant odor or taste, alter the solubility, and prevent evaporation of the substances or other incompatibilities (Baysan et al., 2019; Pai et al., 2015; Şahin-Nadeem et al., 2013). There are many different encapsulation methods for bioactive compounds, but spray drying is an industrial and economical method commonly used to transform liquid products into dry powders (Chew et al., 2018; Li et al., 2017; Murugesan & Orsat, 2012).

Previous studies have revealed that the wall properties determine the encapsulation efficiency (EE) (Belščak-Cvitanović et al., 2015; Chew et al., 2018; Şahin-Nadeem et al., 2013). Therefore, choosing suitable wall materials is a crucial step for efficiently producing encapsulated powders. Individually, β-cyclodextrin (BCD) can be used as an effective carrier for naringin, the most prominent bioflavonoid in citrus fruits. Owing to their unique

Received 03 Oct., 2021
Accepted 19 Nov., 2021
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structure and lipophilic surface, BCD molecules can easily form inclusion complexes with a wide variety of molecules and molecular ions, thus enhancing the solubility and bioavailability of the compounds (Cui et al., 2012). However, the high cost of BCD is the major limitation to its use, and resistant maltodextrin (RMD) is a potential replacement that can encapsulate and enhance the solubility of naringin (Pai et al., 2015). RMD has a low glycemic index (10% that of maltodextrin) and is used as an indigestible fiber (with almost 40% fermented into short-chain fatty acids). It also has several nutritional benefits as a prebiotic (Pai et al., 2015).

In some cases, when a single encapsulating matrix cannot fulfill all the required characteristics, better results can be achieved with a mixture of multiple wall materials (Baranauskaite et al., 2019; Chew et al., 2018; Vo et al., 2019). For avocado seed phenolic compounds, a mixture of 20:80 maltodextrin (MD) and gum arabic (GA) was selected as a coating agent instead of using a single coating agent (Vo et al., 2019). The combination of MD and pectin was also used to mask the unpleasant smell of herbal extracts, thus making them suitable for successive manufacturing to produce functional components of food or for nutraceuticals purposes (Sansone et al., 2011). Gelatin supplemented with GA, Tween 20, and BCD was also used to microencapsulate the Turkish oregano extract (Baranauskaite et al., 2019).

The objective of the present work was to optimize the wall material composition (RMD, pectin, BCD, and GA) of pomelo microcapsules prepared by spray drying. We employed the response surface methodology (RSM) to analyze the two response variables of polyphenol recovery yield (PRY) and EE to maximize their synergistic effect.

2 Materials and methods

2.1 Materials and chemicals

Pomelo peels of Da Xanh cultivar were collected from facilities that focused on minimal processing, located in the Mekong Delta of Vietnam. The facilities ensured that the fruits originated from Ben Tre provinces (Vietnam), where they have been granted a geographical indication by the local government.

RMD (resistant starch type V, GI < 10) was provided by the Essential Flavours Vietnam Company (Long An city, Vietnam). Pectin (from apple, DE 70-75%) was purchased from Sigma-Aldrich (USA). BCD was supplied by Shanghai Zhanyun Fine Chemical Co., Ltd. (Shanghai, China). GA was supplied by HiMedia (Mumbai, India). Ethanol extraction solvent (99.5% v/v) was purchased from CEMACO (Ho Chi Ming city, Vietnam).

All other chemicals were of high-purity analytical grade.

2.2 Extracting polyphenol from pomelo peel

This study used a particular extraction procedure that was designed to recover water-soluble phenolic fractions from the pomelo peel of Da Xanh cultivar, according to the method given by To & Nguyen (2021). Fresh pomelo peel was shredded into pieces (size < 1.5 mm), soaked in an equal amount of 40% ethanol solution (80-90 °C), and heated to 85-90 °C for 2 min. The mixture was then pressed, and the liquid fraction was recovered. Ethanol was removed from the solvent using rotary evaporation equipment (HeiVAP Precision (ML), Heidolph Instruments Co., Ltd., Schwabach, Germany), which set the absolute pressure at 160-180 mbar and the temperature of the water bath at 65 °C. The process was stopped when approximately 85% of the total weight was lost. The obtained extract was then added to 1% diatomite (w/w) and filtered through the Whatman filter paper (grade 4: 20-25 μm). This step removed the precipitation that appeared when ethanol evaporated. At this time, the soluble solid of the concentrated extract was approximately 30-40 °Bx; therefore, distilled water was added to standardize it to 30 °Bx.

The final concentrated extract was stored frozen at –18 °C for further use.

2.3 Microencapsulation by spray drying

The pomelo peel extract was thawed by heating at 30 °C. Phenolic compounds in the peel extract were then microencapsulated by spray drying. Following the experimental design, a known amount of each wall material was mixed with 50 g of the pomelo peel extract. The mixture was then stirred, diluted with water to 30 °Bx, and homogenized at 7513 g for 5 min (Pasrija et al., 2015). The mixture was preheated to 60 °C before being loading into a laboratory-type spray dryer (DHSL.SD303; DHSLKOREA, Seoul, Korea). The spray drying inlet and outlet temperatures were fixed at 180 °C and 65 °C, respectively, according to Baysan et al. (2019). The obtained microcapsules were collected from the collector and taken directly into the capped sample vessels, which were stored at 4 °C in the dark before analysis.

2.4 Polyphenol content analysis

The Folin-Ciocalteu assay was employed to determine the polyphenol content, according to the method given by Siddiqua et al. (2010) with minor modifications. The liquid containing polyphenol (0.1 mL) was mixed with 1.5 mL of 1:9 diluted Folin-Ciocalteu's phenol reagent, followed by 4 mL of sodium carbonate (20%, w/v), and distilled water to achieve a total volume of 10 mL. The mixture was allowed to stand for 30 min in the dark at room temperature, and then the absorbance was read at 738 nm using a spectrophotometer (Spectroquant® Pharo 300, Merck Millipore, Darmstadt, Germany). The polyphenol content was calculated as mg of gallic acid equivalents per gram of dry matter from a standard curve of gallic acid. To determine the total polyphenol content (TPC) and surface polyphenol content (SPC), 1 g of microcapsules was dissolved in 9 mL of distilled water (To & Nguyen, 2021) or 9 mL of ethanol:methanol (50:50, v/v) (Vo et al., 2019), respectively. The polyphenol content of the liquid/supernatant was quantified as described above. For the theoretical polyphenol content (TPC_t), the theoretical value was calculated based on the polyphenol content of the concentrated extract using the material balance equation. The concentrated extract was diluted ten times before quantification.

PRY was calculated from TPC and TPC_t as follows (Equation 1):

$$PRY(\%) = \frac{TPC}{TPC_t} x100\% \quad (1)$$

EE was calculated from TPC and SPC as follows (Equation 2):

$$EE(\%) = \frac{TPC - SPC}{TPC} x100\% \quad (2)$$

2.5 Experimental design and statistical analysis

RSM was used to predict the composition of the wall materials to microencapsulate phenolic compounds from pomelo peel. The Box-Behnken design with a second-order polynomial equation was selected to investigate the combined effects of the four independent variables. The independent variables listed were RMD (X_1, %), pectin (X_2, %), BCD (X_3, %), and GA (X_4, %). The ranges of each variable and central points were selected based on the preliminary experimental results. The coded and uncoded independent variables used in the RSM design are listed in Table 1. Table 2 presents the uncoded experimental design scheme and response values of PRY (Y_1%) and EE (Y_2%). The experimental data were fitted to a quadratic polynomial model, and regression coefficients were obtained. The quadratic model used for the response surface is as follows (Equation 3):

$$Y = \alpha_0 + \sum_{i=1}^{4} \alpha_i X_i + \sum_{i \neq j=1}^{4} \alpha_{ij} X_i X_j \quad (3)$$

where Y denotes the dependent variable, a_0 is a constant, and a_i, a_{ii}, and aij are the linear, quadratic, and interactive coefficients of the model, respectively. X_i and X_j are the levels of independent variables.

Three-dimensional surface response plots were generated by varying two variables within the experimental range and holding the other constants at the central points. The quality of the fitted model was expressed by the coefficient of determination (R^2) and the adjusted coefficient of determination (R^2_{adj}), and the statistical significance of the model was determined by the F-test. Mathematical analyses were conducted using Statgraphics Centurion v.19 (Statgraphics Technologies, Inc. Computer Software, The Plains, Virginia). The test of statistical significance was based on the total error criteria, with a confidence level of 95.0%.

3 Results and Discussion

3.1 Fit-to-model

In this study, RSM was employed to analyze two response variables: PRY and EE. The results obtained from the 30 runs are listed in Table 2. These experimental data were used to calculate the coefficients of the second-order polynomial equations.

Table 1. Uncoded and coded levels of independent variables used in the response surface methodology (RSM) design.

Symbols	Independent variables	Coded levels		
		−1	0	1
X_1	Resistance maltodextrin (%)	45.0	52.5	60.0
X_2	Pectin (%)	0.5	1.0	1.5
X_3	β-cyclodextrin (%)	3.0	5.0	7.0
X_4	Gum arabic (%)	4.0	6.0	8.0

Table 3 summarizes the results, including regression coefficients,the significance of the coefficients of the models (F-value and P-value, by ANOVA), and the coefficient of determination. In general, a small P-value indicates a more significant effect on the respective response variables. By excluding all of the interactions that poorly fit the models ($P > 0.05$), the accuracy of the model can be greatly improved. In reality, the results of ANOVA showed that the obtained second-order polynomial models fit well with the observed data. The R^2 and R^2_{adj} values for the response of PRY were 0.9962 and 0.9993, respectively. Meanwhile, for the response of EE, the R^2 was 0.9983, and the R^2_{adj} value was 0.9972. In both cases, R^2 was in reasonable agreement with R^2_{adj}.

3.2 Response surface analysis of PRY

By performing multiple regression analysis on the experimental data, the changes in the dependent variable PRY (Y_1) could be expressed by a second-order polynomial model, as shown in Equation 4. In this relationship, the interaction between pectin and BCD was not significant. As mentioned, a high coefficient of determination (R^2=0.9962) indicated a good fit.

$$\begin{aligned} Y_1 = & -313,926 + 10,2426.X_1 + 52,99.X_2 + 19,621.X_3 + \\ & 13,4879.X_4 - 0,0968741.X_1^2 - 0,32.X_1.X_2 - 0,105.X_1.X_3 + \\ & 0,145167.X_1.X_4 - 23,6717.X_2^2 + 1,585.X_2.X_4 - \\ & 1,45635.X_3^2 + 0,22375.X_3.X_4 - 1,88167.X_4^2 \end{aligned} \quad (4)$$

To aid in visualization, 3D response surfaces were generated from the model and are graphically represented in Figure 1. The same trends were observed for the four wall materials, in which all showed positive linear effects but negative quadratic effects on PRY. Excluding the interaction between pectin & BCD, the two interactions between RMD and pectin, RMD and BCD showed positive effects; however, the three interactions between RMD and GA, pectin and GA, BCD and GA showed negative effects. The combination of all these opposite factors resulted in parabolic effects on PRY, meaning that too low or too high of each wall material could also cause PRY to decrease. Therefore, the maximal Y_1 was predicted to be 79.00% when the wall material composition consisted of 53.15% RMD, 0.97% pectin, 5.31% BCD, and 6.36% GA, which is very close to the central point (52.5%, 1.00%, 5.00%, and 6.00%, respectively).

3.3 Response surface analysis of EE

Similarly, multiple regression analysis was performed on the experimental data of the dependent variable EE (Y_2). A second-order polynomial model can also be used to express the change in Y_2, as shown in Equation 5. However, the interactions between pectin and BCD, and BCD and GA, were not significant in this case. A high coefficient of determination (R^2=0.9983) was also obtained, indicating a good fit.

Figure 2 represented the 3D response surfaces generated from the model. The same trends were also observed, in which

Table 2. Experimental design and the responses of dependent variables to wall material composition.

No.[a]	Independent variable				Dependent variable		
	RMD	Pectin	BCD	Gum arabic	PRY	EE	Observed desirability
	X_1 (%)	X_2 (%)	X_3 (%)	X_4 (%)	Y_1 (%)	Y_2 (%)	
1	60.0	1.5	5.0	6.0	66.37	71.64	0.3516
2	52.5	1.0	3.0	4.0	62.59	68.54	0.0627
3	45.0	1.0	5.0	8.0	64.87	73.02	0.3672
4	60.0	1.0	7.0	6.0	68.03	73.40	0.4859
5	45.0	1.5	5.0	6.0	67.54	72.70	0.4378
6	52.5	1.5	7.0	6.0	68.24	73.20	0.4823
7	45.0	1.0	3.0	6.0	63.33	71.12	0.2372
8	52.5	1.0	5.0	6.0	78.58	77.02	0.9587
9	60.0	1.0	5.0	4.0	61.65	68.29	0.0000
10	52.5	0.5	3.0	6.0	65.94	71.85	0.3504
11	52.5	0.5	5.0	8.0	67.22	73.90	0.4834
12	60.0	1.0	3.0	6.0	67.88	71.71	0.3939
13	60.0	1.0	5.0	8.0	70.95	75.25	0.6598
14	52.5	1.0	3.0	8.0	65.22	72.78	0.3698
15	52.5	1.0	7.0	4.0	63.93	71.36	0.2671
16	52.5	1.0	5.0	6.0	78.32	77.28	0.9661
17	52.5	1.0	5.0	6.0	78.88	77.64	1.0000
18	52.5	1.0	5.0	6.0	78.58	77.26	0.9717
19	45.0	1.0	7.0	6.0	69.78	74.54	0.5916
20	52.5	1.0	5.0	6.0	78.58	77.28	0.9728
21	52.5	0.5	7.0	6.0	69.32	74.32	0.5676
22	52.5	0.5	5.0	4.0	64.96	71.02	0.2814
23	45.0	0.5	5.0	6.0	66.14	72.93	0.4065
24	52.5	1.0	5.0	6.0	78.59	77.28	0.9731
25	45.0	1.0	5.0	4.0	64.28	71.30	0.2754
26	60.0	0.5	5.0	6.0	69.77	73.40	0.5347
27	52.5	1.0	7.0	8.0	70.14	75.20	0.6331
28	52.5	1.5	3.0	6.0	63.18	70.70	0.2142
29	52.5	1.5	5.0	4.0	59.78	68.70	0.0000
30	52.5	1.5	5.0	8.0	68.38	74.21	0.5339

[a]Experiments were conducted in a random order.

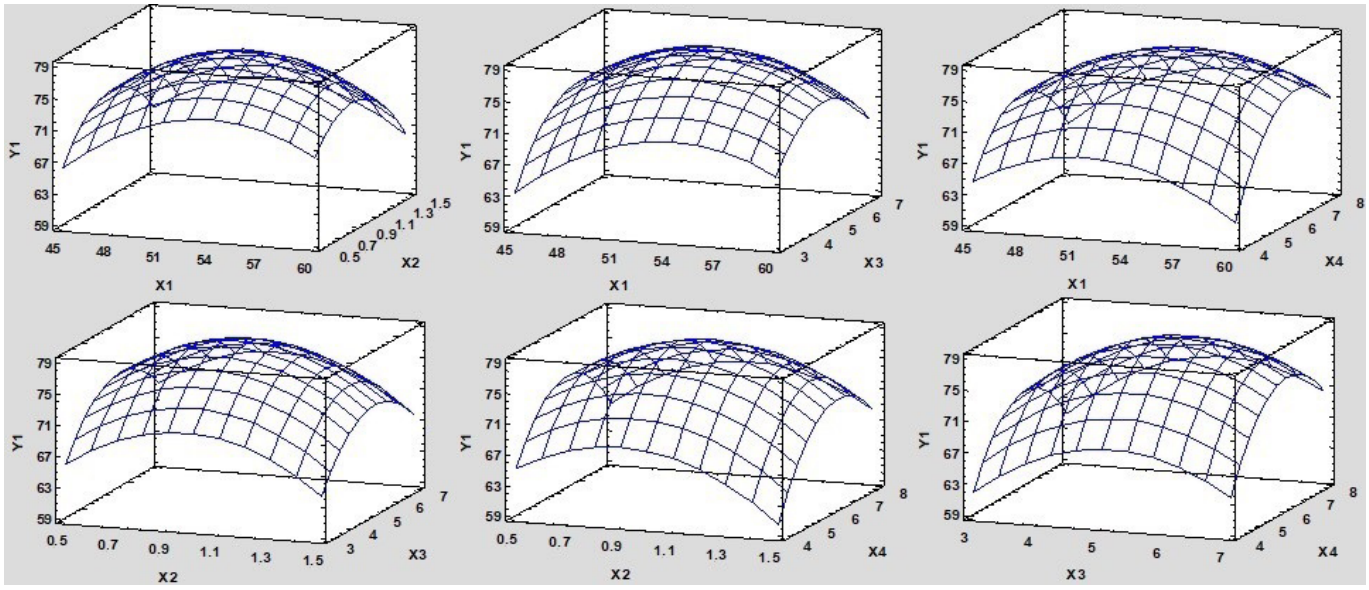

Figure 1. Surface plots of polyphenol recovery yield (PRY) (Y_1,%) as a function of the concentrations of resistant maltodextrin (RMD) (X_1, %), pectin (X_2,%), β-cyclodextrin (BCD) (X_3, %), and gum arabic (GA) (X_4,%).

Table 3. Regression coefficients of the predicted second-order polynomial models for polyphenol recovery yield (PRY) and encapsulation efficiency (EE).

	Polyphenol recovery yield			Encapsulation efficiency		
	Regression coefficients	F-value	P-value	Regression coefficients	F-value	P-value
a_0	−313.926			−743.071		
Linear						
a_1	102.426	28.72	0.0001	395.528	15.12	0.0012
a_2	52.99	36.80	0.0000	193.283	161.21	0.0000
a_3	19.621	171.75	0.0000	803.125	962.45	0.0000
a_4	134.879	331.46	0.0000	479.167	2593.81	0.0000
Quadratic						
a_{11}	−0.0968741	924.96	0.0000	-0.0405185	1752.82	0.0000
a_{22}	−236.717	1090.93	0.0000	−948.167	1895.99	0.0000
a_{33}	−145.635	1057.10	0.0000	−0.587917	1866.11	0.0000
a_{44}	−188.167	1764.68	0.0000	−0.748854	3027.61	0.0000
Interaction						
a_{12}	−0.32	26.17	0.0001	−0.102	28.80	0.0001
a_{13}	−0.105	45.08	0.0000	-0.0288333	36.82	0.0000
a_{14}	0.145167	86.16	0.0000	0.0873333	337.79	0.0000
a_{23}		*(p > 0.05, excluded)*			*(p > 0.05, excluded)*	
a_{24}	1.585	45.65	0.0000	0.6575	85.09	0.0000
a_{34}	0.22375	14.56	0.0015		*(p > 0.05, excluded)*	
R^2	0.996214			0.998345		
R^2_{adj}	0.993137			0.997176		

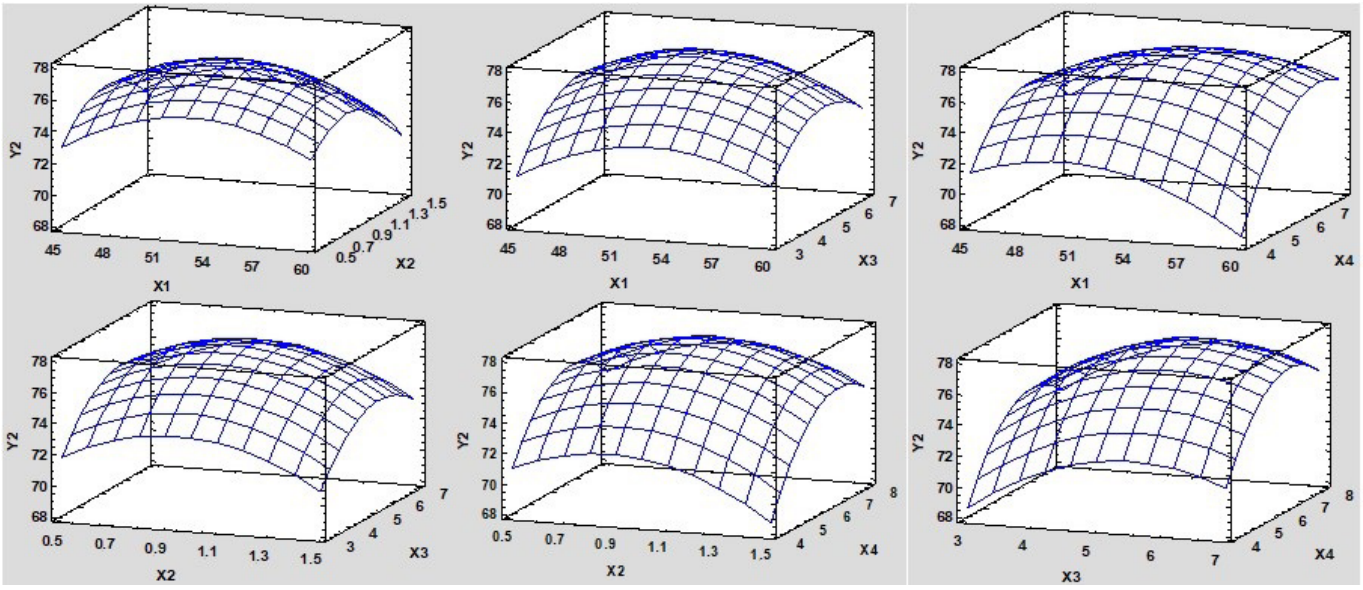

Figure 2. Surface plots of encapsulation efficiency (EE) (Y_2, %) as a function of the concentrations of RMD (X_1, %), pectin (X_2, %), BCD (X_3, %), and GA (X_4, %).

all four independent variables showed positive linear effects but negative quadratic effects on PRY. The two interactions between RMD and pectin, RMD and BCD showed positive effects, and in contrast, the two interactions between RMD and GA, pectin and GA, showed negative effects. This means an addition of walls material had positive effects on EE (Y_2) and can increase its value at a lower concentration. However, for a higher amount, the negative quadratic effect was significant, thus causing the decrease in EE. These are the same parabolic effects observed on PRY. In this case, the maximal Y_2 was predicted to be 77.84% when wall materials consisted of 52.85% RMD, 0.97% pectin, 5.53% BCD, and 6.7% GA, respectively. This formula was lower in RMD and higher in BCE and GA in comparison with the one of Y_1.

$$Y_2 = -74,3071 + 3,95528.X_1 + 19,3283.X_2 + 8,03125.X_3 + 4,79167.X_4 - 0,0405185.X_1^2 - 0,102.X_1.X_2 - 0,0288333.X_1.X_3 + 0,0873333.X_1.X_4 - 9,48167.X_2^2 + 0,6575.X_2.X_4 - 0,587917.X_3^2 - 0,748854.X_4^2 \quad (5)$$

3.4 Optimization of the wall material composition for spray drying

The spray drying conditions would be optimum if Y_1 and Y_2 reached maximum values simultaneously. Thus, multiresponse optimization methods were employed. In this method, the dependent variables were converted to desirability values, which ranged from 0 to 1, from minimum to maximum. The calculation was carried out using Statgraphics Centurion XV software, and all the responses were converted to desirability values and are presented in Table 2. At the same time, the most effective wall material compositions for PRY and EE were generated by optimizing the desirability function. Figure 3 shows the overlay contour plots of the optimization of the four wall materials for the two response variables Y_1 and Y_2. The predicted optimum values of desirability value were 1.00, when 52.66% RMD, 0.92% pectin, 5.30% BCD, and 6.28% GA were used. This desirability corresponded with the values of PRY at 78.90% and EE at 77.67%. The suitability of the model was verified by additional independent experiments in which the microencapsulation process was carried out with the optimal formula in triplicate.

The experimental values of PRY and EE were 78.86% and 77.78%, respectively (Table 4). These values matched well with those predicted by the regression models.

4 Discussion

As mentioned above, phenolic compounds are sensitive to physicochemical factors. Spray drying at high temperature and high air velocity made phenolic compounds more susceptible to degradation, which is the cause of the decrease in the total phenol content of the extract, thus leading to the reduction of PRY. Meanwhile, the EE reflects the potential of the wall material to hold the core material inside the microcapsule. A high EE always indicates a high retention of the core material in the wall material. In this study, the concentration of each wall material had parabolic effects on PRY and EE, meaning that too low or too high of each carrier could also cause PRY and EE to decrease.

An increase in PRY and EE has been observed in many studies. Pai et al. (2015) reported that 60% to 80% encapsulation of naringin was achieved when RMD loading increased from 20% to 40% (w/v). The increase in EE is most probably due to the increase in the thickness of the microparticle shell walls. Naringin is considered a poorly soluble drug, has a tendency to undergo phase separation and stick to the dryer surface, thus resulting in inferior EE. Therefore, increasing the RMD loading possibly increased the EE due to the reduced phase separation. Similarly, Sidlagatta et al. (2020) also reported the increase of process yield of concentrated orange juice (from 21.35 to 56.95%) when increased maltodextrin concentration of the feeding liquid (from 1:0.5 to 1:1.5, total soluble solid to maltodextrin level). However, this study did not analyze the polyphenol content of the juice. Sansone et al. (2011) selected 10% MD and 1% feed concentration for spray-drying nutraceutical extracts. When the maltodextrin concentration was lower than 10%, most materials were stuck on the spray dryer chamber wall, reflecting a low production yield. Meanwhile, pectin concentrations lower than 1% were not able to produce well-formed and completely coated microparticles. In addition, the selection of carrier materials in the right proportion also played an important role (Belščak-Cvitanović et al., 2015; Chew et al., 2018; Vo et al., 2019). In assessing natural biopolymers as encapsulants of green tea bioactive compounds, Belščak-Cvitanović et al. (2015) also reported that a lower carrier concentration provides more inferior polyphenol entrapment, while higher concentrations

Table 4. Experimental data of verification of the wall material composition.

	Experimental value				Predicted value
	1st	2nd	3rd	Mean ± SD	
PRY (Y_1, %)	78.84	79.01	78.73	78.86 ± 0.14	78.90
EE (Y_2, %)	77.75	77.70	77.88	77.78 ± 0.09	77.67

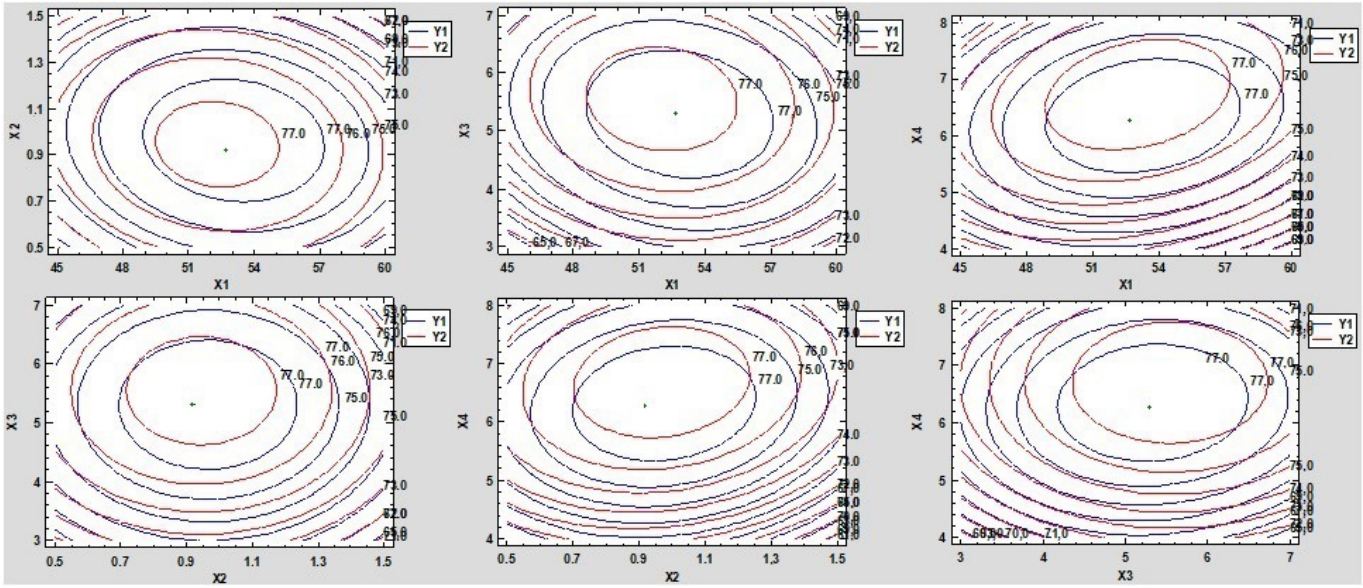

Figure 3. Overlay contour plots of PRY (Y_1) and EE (Y_2) as a function of the concentrations of RMD (X_1), pectin (X_2), BCD (X_3), and GA (X_4).

of the carrier were more efficient in protecting and entrapping polyphenols. According to the obtained results, only the use of modified starch and inulin as the carriers of green tea bioactive compounds provided significantly higher loading capacities of both TPC and flavan-3-ols (EE of TPC was 82.16% and of flavan-3-ols was 75.94% in the case of inulin, 80.03% and 82.25% in the case of modified starch, respectively). Of the remaining ten carriers, only pectin and alginate retained higher but insignificant polyphenol and flavan-3-ols content. The beneficial and protective effects of inulin, modified starch, alginate, and pectin may be explained by the interactions between these polysaccharide materials and polyphenolic compounds. For avocado seed phenolic compounds, Vo et al. (2019) proposed using a mixture of 20:80 MD:GA ratio as a coating agent for encapsulation. Of the six different combinations, a mixture of 20:80 MD:GA ratio increased PRY to 98.6% (from 78.0% when using GA only and 79.2% when using MD only). The importance of the selection of wall materials was also implied in the microencapsulation of pequi oil, (Santos et al., 2020) and kenaf seed oil (Chew et al., 2018). Santos et al. (2020) recommended using a 25:75 mixture of GA and MD as wall material for the microencapsulation of pequi oil to achieve smaller droplet size and lower zeta potential. Chew et al. (2018) considered BCD unique in terms of its non-polar cavity in the interior surface and a hydrophilic exterior surface that contributes to the capability of a host-guest inclusion complex. It was theorized that the wall properties might be improved by combining with different agents and possess all the qualities required for ideal encapsulation. The results also proved that the carbohydrate-based microcapsules (BCD mixed with GA) achieved the highest EE (95.3%). In addition to kenaf seed oil, the interaction of BCD with naringin, the most prominent bioflavonoid in Citrus fruit, has also been reported by Cui et al. (2012).

However, the decrease in PRY and EE by increasing the concentration of the carriers was not observed in many studies, including those by Pai et al. (2015) and Belščak-Cvitanović et al. (2015). Similar results were also seen in the impact of gelatin supplemented with GA, Tween 20, and BCD on the microencapsulation of Turkish oregano extract reported by Baranauskaite et al. (2019). By using D-optimal design, the optimized wall material solution was predicted to consist of 10 g of gelatin, 7.5 g of GA, 1.99 g of Tween 20, and 1.98 g of BCD (to 20 g of ethanolic oregano extract), which was very close to the upper limit of the model (10 g, 7.5 g, 2 g, and 2 g respectively). In contrast, Şahin-Nadeem et al. (2013) determined that the loading capacity of phenolic compounds of the sage extract in maltodextrin or GA microparticles was significantly decreased by increasing the concentration of the carriers, which may be associated with the dilution of the sage extract by the addition of carrier material. According to the increasing concentration of carriers, Sansone et al. (2011) also reported a decrease in production yield. Maltodextrin concentrations higher than 10% resulted in products showing the presence of agglomerations and powder cakes, as well as low retention of the core material. This behavior was probably due to a delay in the formation of a semi-permeable layer by the internal components during drying. Meanwhile, pectin concentrations higher than 1% enhanced the viscosity of the liquid feed, which is not suitable for processing, preventing the proper formation of droplets during the atomization stage of the spray drying process. Xu et al. (2019) used only GA for the microencapsulation of mulberry polyphenols. When the core/wall ranged from 1:50 to 1:110, it was reported that the ratio of 1:90 achieved the highest EE (EETPC = 95.54%; EETFC = 97.26%; EETAC = 97.36%). This could be interpreted from the following aspects: On one hand, excessive GA content leads to high viscosity that decreases the EE; on the other hand, excessive core material reduces the strength of the wall material, thus decreasing the EE.

5 Conclusions

In this study, RSM was employed to predict the optimum wall material composition for the microencapsulation of phenolic compounds from pomelo peels. Using a graphical optimization method, which combined RSM with a desirability function, the optimal wall material composition was predicted to consist of 52.66% RMD, 0.92% pectin, 5.30% BCD, and 6.28% GA. In this case, the expected PRY and EE were 78.86% and 77.78%, respectively. These values are consistent with the experimental values, thus proving the reliability of the obtained model.

Acknowledgements

The authors would like to acknowledge the financial support from the Vietnam Ministry of Education and Training under project code B2020-TCT-01.

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