OPTIMIZATION OF SPRAY DRYING PROCESS OF GINGER OLEORESIN USING RESPONSE SURFACE METHODOLOGY

TRUONG THI QUYNH TRAM[*], DANG QUOC TUAN[**]

ABSTRACT

In this study, response surface methodology was used to establish optimum conditions for microencapsulation of ginger oleoresin. The best microencapsulation was obtained using a combination of 20.7% maltodextrin, 4.2% gelatin, 10.0% oleoresin loading. This condition resulted in high microencapsulation efficiency (88.1%) and low moisture content (3.9%).

Keywords: ginger oleoresin, response surface methodology, spray drying.

TÓM TẮT

Tối ưu hóa quá trình sấy phun nhựa dầu gừng bằng phương pháp bề mặt đáp ứng

Trong nghiên cứu này, phương pháp bề mặt đáp ứng đã được sử dụng nhằm khảo sát vùng điều kiện tối ưu cho quá trình sấy phun nhựa dầu gừng (ginger oleoresin). Điều kiện sấy phun tối ưu nhất đạt được với giá trị của các biến 20.7% maltodextrin, 4.2% gelatin và 10.0% oleoresin;điều kiện này cho hiệu suất sấy phun là 88.1% và độ ẩm bột là 3.9%.

Từ khóa: nhựa dầu gừng (ginger oleoresin), phương pháp bề mặt đáp ứng, sấy phun.

1. Introduction

Ginger oleoresin is an extract from ginger (*Zingiber officiale* Roscoe) which has a flavor profile approaching the ground fresh spice [8]. Oleoresin contains the nonvolatile pungent principles of ginger in addition to some essential oils and other nonvolatile compounds such as fixed oil, resin, fatty acids, and pigments [10]. Extraction yield of ginger oleoresin has been reported to be in the range of 3.5-10%. The variation of oil content is depending on the ginger varieties, age of harvest, and extraction conditions [8]. For food use, oleoresin is more desirable than ground spice because the former is hygienic and can be standardized for acceptable flavour levels by blending [5]. In comparison with the essential oils, oleoresin contains natural antioxidants of the corresponding spices, which make them more stable [6]. It provides better distribution in the finished products and requires less storage space than the corresponding spices [9]. However, ginger oleoresin is susceptible to degradation by high temperatures and the presence of oxygen and light. Processing conditions of foods can cause degradation of ginger oleoresin reducing its functional properties [3]. It can

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also react with components present in the food system, which may limit bioavailability, or change the color or taste of food [10]. In many cases, microencapsulation has been widely adopted as an approach to address this issue.

Microencapsulation is defined as a process in which an active compound is covered with a protective wall material [7]. The microcapsules may range from 0.2 to 5000 µm in size and have multitudes of shapes, depending on the materials and methods used to prepare them. Using the appropriate encapsulating substances, core component in microcapsules can be protected from damage caused by adverse environmental conditions such as light, moisture and oxygen [7]. The encapsulated materials are prevented from degradation reactions, loss of aroma, and thus maintain their stability [8]. In addition, in the form of powder, handling and use of the active ingredients into food and beverages becomes easier.

Characteristic of coating material is one of the most important factors that affect the microencapsulation process and its product [9]. The commonly used coating materials are gum Arabic, maltodextrin, modified starch, protein, gelatin, and glucose syrup; each has its advantages and disadvantages [6]. The right combination of coating materials will produce encapsulated product with desired characteristics. Gum Arabic yields a stable emulsion with most oils over a wide pH range and forms a visible film at the oil interface. However, price and availability of the materials limit the use for encapsulation purposes [7]. Maltodextrin has been studied as a replacer of gum Arabic in spray-dried emulsion [3]. Maltodextrin is a starch derivative that can reduce deposit product attached to the dryer wall. However, maltodextrin exhibits poor emulsifying capacity, emulsion stability and low oil retention [9]. Mixture of maltodextrin and sodium caseinate was reported to be effective in microencapsulation of ginger oleoresin using spray drying [3]. Gelatin, when compared to maltodextrin, possesses all the properties of an effective entrapping agent: high emulsifying activity, high stabilizing activity, and a tendency to form a fine dense network upon drying [7]. The screening of polymer blends that could result in higher encapsulating efficiency and lower cost than the individual biopolymers has been object of increasing interest [3]. The most common and economical technique used for micro-encapsulation process is the spray drying [8]. Microencapsulation by spray drying has been successfully used in the food industry for several decades [7]. This technique provides a high retention of aroma compounds during drying. Spray drying enables the transformation of feed from a fluid state into dried particulate form by spraying the feed into a hot drying medium. There are very few reports on the optimization of ginger oleoresin using spray drying [3]. Microencapsulation of black pepper oleoresin by spray drying, using gum Arabic and the commercial modified starch (e.g., Hi-Cap) as coating materials, has been described [9]. Response surface methodology has been applied in various studies of food such as optimization of microencapsulation of flaxseed oil [6], and optimization of microencapsulation of sunflower oil by spray dying [1]. Box-Behnken design, a spherical and revolving design, has been widely used and demonstrated to be more efficient than the central composite design [2].

In the light of indicated information, an attempt was made to microencapsulate of ginger oleoresin using spray drying with the microencapsulation condition of ginger oleoresin using response surface methodology (RSM) was reported.

2. Materials and methods

2.1. Materials

Fresh ginger rhizome of 7 month age was collected from a local farm in Dong Nai province. The rhizome was cleaned, washed and sliced into 2 mm slices. Then the rhizomes were subjected to blanching at 90°C for 3 minutes to inactivate enzymes. The water content was reduced to reach the final moisture about 8-10% by oven drying at 60°C for 24h. The dried rhizomes were milled and soaked into ethanol 96% to extract the oleoresin. After the extraction period, the extract was filtered and concentrated using a rotary vacuum evaporator (Rotavapor) at 50°C temperature until all solvent was removed for quantification of the dried extract. The extract was dissolved back into ethanol, stored in a flask. This oleoresin was then used as an active ingredient in the micro-encapsulation [3].

Maltodextrin 12 DE (Roquette, France) and gelatin with a bloom value of 260 (Rousselot, France) were used as coating materials. A suspension of coating materials was prepared by mixing a solution of maltodextrin in a solution of gelatin with different ratios of maltodextrin and gelatin in distilled water. The gelatin and maltodextrin were dissolved separately in warm distilled water at 60°C using a homogenizer at low speed (100 rpm). After mixing two components of coating suspension, the suspension was hydrated for 18 hours at room temperature [9].

2.2. Experimental design for response surface methodology

The experimental design chosen for this study was Box Behnken, a fractional factorial design for three independent variables at three levels [4]. This design is preferred because relatively few experimental combinations of the variables are adequate to estimate complex response functions. To determine the optimal conditions for microencapsulation of ginger oleoresin, the effects of three variables (i.e., ginger oleoresin concentration, maltodextrin concentration, and gelatin concentration) on the microencapsulation efficiency were investigated and analyzed systematically by Box-Behnken design as shown on table 1. According to the principle of Box-Behnken design [1], maltodextrin concentration, gelatin concentration and ginger oleoresin concentration were taken as the variables tested in a 16-run experiment including 4 center point to determine their optimum levels in order to achieve the criteria of minimum moisture content and maximum microencapsulation efficiency. All experiments were performed in triplicate and the averages of microencapsulation efficiency and moisture content were taken as response. Experimental data were statistically analyzed by Design-Expert version 7.0.3 (State-Ease, Inc., Minneapolis MN, USA).

The quadratic response surface analysis was based on multiple linear regressions taking into account linear, quadratic and interaction effects according to the equation below:

$$Y= b_0+\sum a_i x_i+\sum a_{ij} x_i x_j+\sum a_{ii} x_i^2$$

Where Y is the response value predicted by the model, b_0 is offset value, a_i, a_{ij}, a_{ii} are main (linear) interaction and quadratic coefficients, respectively.

The adequacy of the models was determined using model analysis; lack-of fit test and coefficient of determination (R^2) analysis. For model to be suited, R^2 should be at least 0.80 for a good fitness of a response model. [1]

Table 1. *Levels and code of variables chosen for Box- Behnken design [1]*

Variables	Symbol		Code levels		
	Uncoded	**Coded**	**-1**	**0**	**+1**
Maltodextrin concentration (%) w/v	X_1	x_1	20	22.5	25
Gelatin concentration (%) w/v	X_2	x_2	2	5	8
Oleoresin concentration (%) w/w	X_3	x_3	10	12.5	15

2.3. Microencapsulation of ginger oleoresin by spray drying

Preparation of ginger oleoresin emulsion was done by adding the ginger oleoresin into suspension of the coating materials at different concentration (based on coating materials), and mixed using a homogenizer at a 6,000 rpm for about 30 minutes. Two drops of Tween 80 were added to aid emulsification [3]

The ginger oleoresin emulsion was then dried using a spray dryer Plant-Lab SD-06 (North Yorkshire, UK) with inlet temperature of 160°C, an outlet temperature of 100°C, a feeding rate of 200 mL/h [3].

2.4. Moisture content (MC) of microcapsules

Moisture content of encapsulated powder was determined gravimetrically by oven drying at 105°C for 6 hours [3].

2.5. Microencapsulation efficiency of spray drying process

The encapsulation efficiency was determined by the fraction of the encapsulated oleoresin over the total quantity of oleoresin. The encapsulated oleoresin could be measured based on the surface oleoresin and the total oleoresin [5].

Microencapsulation efficiency (ME) = (Total oleoresin - Surface oleoresin) x 100% / Total oleoresin

Surface oleoresin was determined through following steps: 5 g of microcapsules were precisely weighted in a beaker and 50 mL of hexane were added and shaken during 15 s at ambient temperature to extract superficial oleoresin. The solvent mixture was then filtered through a filter paper, and after that, the un-encapsulated oleoresin was obtained after vacuum evaporation of hexane [5]. For total oleoresin determination: 5 g of microcapsules were precisely weighted in a beaker and 50 mL of hexane were added to the same powders and the mixture was mixed for 4h. After filtering through a filter paper, hexane was evaporated from the filtrate and total oleoresin was weighed [5].

2.6. Microcapsules morphology

The external appearance (shape and size) of ginger oleoresin microcapsules was determined by a JSM-6480 LV scanning electron microscope (JEOL Company, Japan). The microcapsules were mounted on specimen stubs with double side adhesive carbon tapes. The specimen was coated with a gold film [1].

3. Results and discussion

3.1. Extraction yield

Commercial dried ginger has been reported to contain oleoresins in the yields of 3.5 -10 %, and the pungent principle accounting to 25 % of the oleoresins [10]. The extraction yield in this study was 9%. This probably due to the residual solvent that is still quite high; the residual solvent contained in the specification of ginger oleoresin in the trade was <10 ppm. The remaining ethanol in the oleoresin evaporated during spray drying process, because it has a relatively low boiling point (around 78^0C). Oleoresins contain the nonvolatile pungent principles of ginger in addition to some essential oils and other nonvolatile compounds such as carbohydrates and fatty acids [10].

3.2. Optimization of microencapsulation of ginger oleoresin

To determine the optimal condition of microencapsulation of ginger oleoresin and the relationship between the responses (MC and ME) and the significant variables, statistical analyses of ANOVA was performed through a joint test of three parameters (Table 2).

The results of ANOVA showed that the quadratic polynomial models were adequately represented experimental data. The coefficients of multiple determination R^2 for the response of moisture content and microencapsulation efficiency were 0.9115 and 0.9608, respectively (Table 2). Results in Table 2 showed the largest effect on moisture content were the quadratic term of maltodextrin, followed by the quadratic term of gelatin, and then followed by the linear term of maltodextrin. While for the ME, the largest effect was from the interaction term between gelatin concentration and oleoresin loading, followed by the quadratic term of maltodextrin, and the linear term of gelatin.

The polynomial model for moisture content (Y_1) and microencapsulation efficiency (Y_2) were regressed by considering only the significant terms and show as below:

$$\mathbf{Y_1 = +61.47569 - 1.30183C^* + 0.093800A^2 + 0.14486B^2}$$

$$\mathbf{Y_2 = + 342.89674 - 4.89800A - 31.54172C + 0.92520AC + 0.24803BC - 0.47310B^2 + 0.41322C^2}$$

** A= Maltodextrin concentration, B= Gelatin concentration, C= Oleoresin concentration*

Table 2. *ANOVA of the regression coefficients of the fitted quadratic equations for MC and ME of the ginger oleoresin powder*

Variables	**Moisture content**			**Microencapsulation efficiency (ME)**		
	Regr. coeff	**F-value**	**P-value**	**Regr. coeff**	**F-value**	**P-value**
a_0	+61.47569	6.87	0.0146	+342.89674	16.36	0.0015
Linear						
A	-4.41433	0.11	0.7481	- 4.89800	21.32	0.0036
B	-0.5882	5.50	0.0574	- 1.21249	3.900E-004	0.9849
C	-1.3018	6.49	0.0437	- 31.54172	15.45	0.0077
Interaction						
AB	-0.0173	0.38	0.5592	+ 0.12620	1.55	0.2599
AC	+0.0240	0.51	0.5025	+ 0.92520	57.76	0.0003
BC	-0.0283	1.02	0.3513	+ 0.24803	5.98	0.0501
Quadratic						
A^2	+0.0938	7.77	0.0317	- 0.18426	2.29	0.1809
B^2	+0.1449	38.43	0.0008	- 0.47310	31.32	0.0014
C^2	+0.0422	1.57	0.2564	+ 0.41322	11.52	0.0146
R^2			0.9115			0.9608
Adj R^2			0.7787			0.9021
Pred R^2			-0.4114			0.3895
Lack of fit		264.36	0.0004		33.39	0.0083

a_0 is a constant, A,B,C are the linear; A^2,B^2,C^2 are the quadratic; and AB,AC,BC are the interactive coefficients of the quadratic polynomial equations, respectively.

Moisture contents varied from 3.63 to 6.08% (wet basis), similar to the results for flaxseed oil powder obtained by spray drying Omar et al. [6]. In general, moisture content increased with increased oleoresin loading and this was significant at $p<0.05$. Gelatin concentrations showed insignificant effect on the moisture content at $p<0.05$ (Fig.7a). The increase in both concentrations of maltodextrin and those of gelatin resulted in the increase in the moisture content of the microcapsules. From Fig.2b and 2c, it can be seen that the lower moisture content can be achieved when the concentration of oleoresin is out of studied range (from 10-15%), but the range of moisture content was typical of the moiture content in microcapsules produced obtained from the spray drying technique (2-6%) [3]. The high water content in the microcapsules was followed by the low storage stability.

ME values were in a range from 71.9 to 90.3%. ME was shown to be strongly influenced by maltodextrin concentrations and oleoresin loading levels at $p<0.05$ in term of linear. Gelatin had shown large effect on ME in term of interaction with oleoresin loading and in term of quadratic.

In general, an increase in the gelatin concentration resulted in the increase in ME values (Figs. 3a, 3b, and 3c). Specifically, when gelatin concentration increased in the range of 2-5%, the ME values increased but further increase of gelatin concentration didn't possitively affect the ME value. The maltodextrin negatively affected on the ME value, the increase of maltodextrin caused the decrease of ME probaly due to its lack of emulsification and low-film capacity.

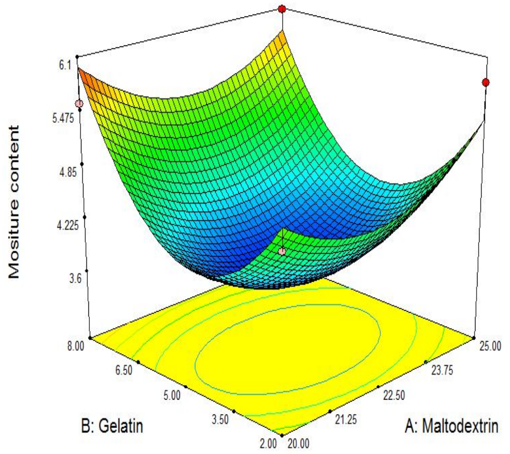

Fig.2a. *Response surface of MC at oleoresin loading of 12.5%*

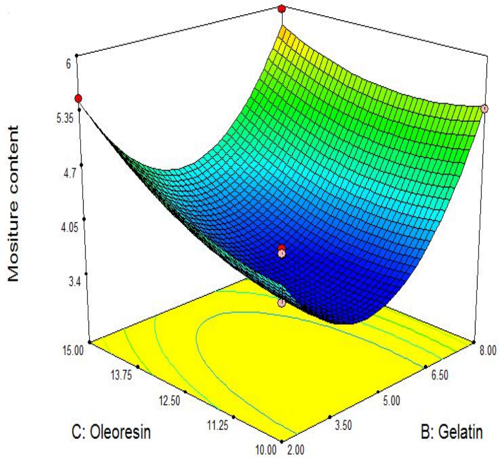

Fig.2b. *Response surface of MC at maltodextrin concentration of 22.5%*

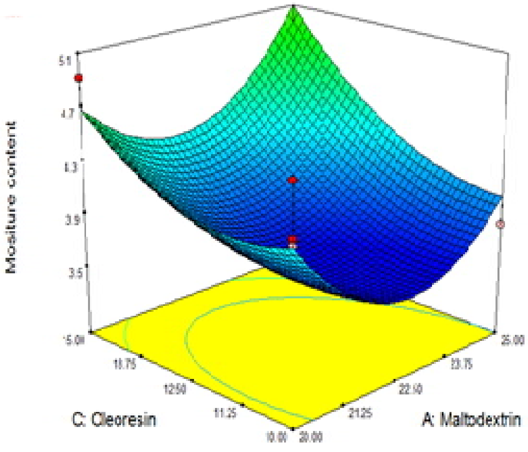

Fig.2c. *Response surface of MC at gelatin concentration of 5%*

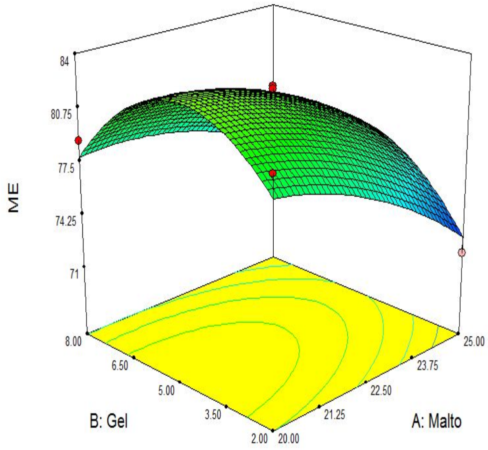

Fig.3a. *Response surface of ME at oleoresin loading of 12.5%*

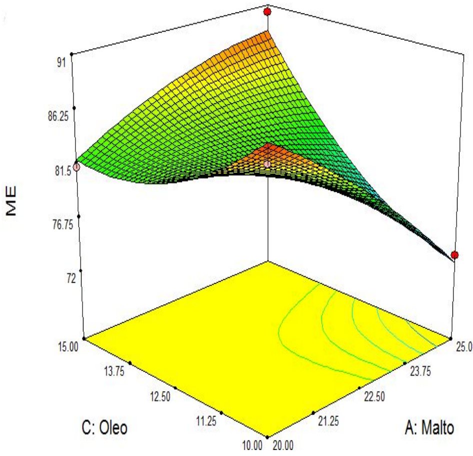

Fig.3b. *Response surface of ME at gelatin concentration of 5%*

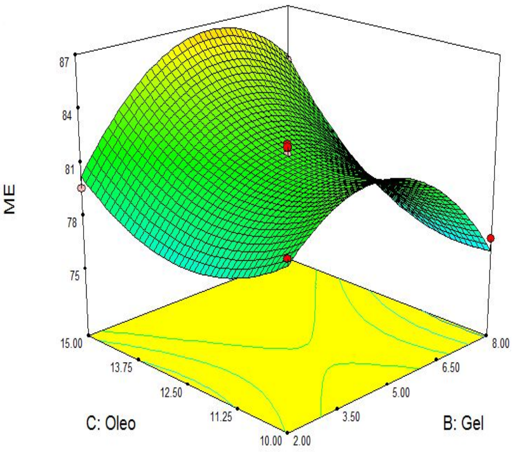

Fig.3c. *Response surface of ME at maltodextrin concentration of 22.5%*

3.3. SEM of microencapsulated ginger oleoresin

SEM micrographs of the spray dried ginger oleoresin produced by the emulsions containing 22.5% of maltodextrin, 2% of gelatin, and 12.5% of ginger oleoresin; and 22.5% of maltodextrin, 8% of gelatin, and 12.5% of ginger oleoresin are shown in Fig.4. Microcapsules prepared from emulsion containing low gelatin concentration had a significant wrinkle on the surface while spherical shape and smooth surface was observed from the microcapsules produced from the emulsion with high concentration of gelatin. The morphology of the microencapsulated ginger oleoresin in Fig. 4 was similar to microcapsules manufactured by the spray-dried microencapsulation [1, 9], implying that MEE can be maximized through optimization of microencapsulation conditions.

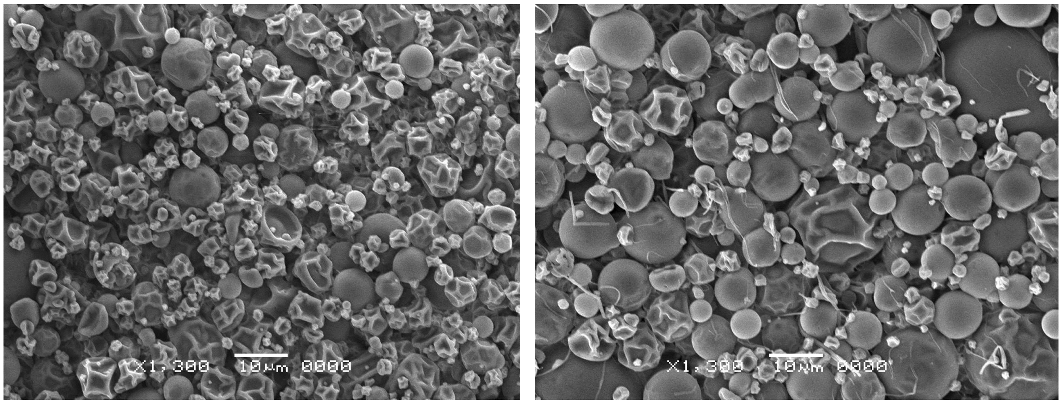

Fig.4. *Spray dried microcapsules of ginger oleoresin in (a) 22.5% of maltodextrin, 2% of gelatin, and 12.5% of ginger oleoresin; and (b) 22.5% of maltodextrin, 8% of gelatin, and 12.5% of ginger oleoresin*

3.4. Optimization procedure

Numerical optimization procedures were carried out for predicting exact optimum level of independent variables leading to desirable response goals. Optimal treatment was found via response surface plotting of the data, by compromise between optimum ranges of the two responses.

The final goal of the optimization process is to produce the microcapsules with high value of the microencapsulation efficiency and the moisture content is in the range of 1-6%. So using response optimizer, the results obtained were 20.7% maltodextrin, 4.2% gelatin, 10.0% oleoresin loading which had shown high value of ME (88.1%) and low moisture content (3.9%).

4. Conclusions

The ginger oleoresin was successfully dried using a lab-scale spray drying apparatus. The study showed that Box-Behnken design was sufficient to describe and predict the responses of the moisture content of microcapsules and microencapsulation efficiency of the system within experimental ranges. The best condition for ginger oleoresin microencapsulation, aiming at achieving high microencapsulation efficiency and low moisture content, were 20.7% maltodextrin, 4.2% gelatin, and 10.0% oleoresin loading which resulted in predicted value of ME of 88.1% and moisture content of 3.9%.

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