Research article

Flavour and Fragrance Journal

Received: 12 February 2015, Revised: 30 March 2015, Accepted: 24 April 2015 Published online in Wiley Online Library: 20 May 2015

(wileyonlinelibrary.com) DOI 10.1002/ffj.3253

Microencapsulation of vanillin by spray drying using soy protein isolate–maltodextrin as wall material

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Abstract: In this study, the application of a response surface methodology (RSM) to optimize the microencapsulation of vanillin as a nutraceutical was studied. Soy protein isolate and maltodextrin were used as a wall material. Microencapsulate of vanillin using spray drying with maltodextrin solutions at different concentration (5–15% wt), vanillin concentrations (0.1–0.4 %wt) and inlet temperature (180–200 °C) were the factors investigated with respect to moisture content, particle size and encapsulation efficiency. RSM was used to determine the optimum processing conditions that yielded maximum encapsulation efficiency and minimum moisture content and particle size during drying of vanillin microencapsulate. The results revealed that microencapsulated powder obtained at 184 °C with a 8.5% maltodextrin concentration and 0.36% vanillin concentration was optimum among investigated samples in terms of its encapsulation efficiency, particle size and moisture content. The scanning electron microscopy (SEM) of the powders produced at the selected optimal conditions showed a spherical shape with no apparent cracks or fissures, which promotes better protection and retention of the core material. Copyright © 2015 John Wiley & Sons, Ltd.

Keywords: microencapsulation; response surface methodology; soy protein isolate; vanillin

Introduction

One of the most popular flavours used in food and beverage is vanillin (4-hydroxy-3-methoxybenzaldehyde). Vanillin is extracted from the seed pods of *Vanilla planifolia*. The addition of this flavouring has multifunctional effects such as antimutagenic, antiangiogenetic, anticolitis, antisickling and antianalgesic.[1,2] In addition, vanillin can increase the concentration of the neurotransmitter serotonin in the brain and increased brain serotonin concentration leads to a reduced craving to consume food.[2] Vanillin's functionality and stability can be improved by its encapsulation into a suitable matrix. In recent years, incorporation of active agents into polymer matrices for extending their shelf life, protecting against oxidation and increasing biological half time have been growing rapidly.[3–6] In this study, we examined the preparation and properties of microencapsulates containing vanillin surrounded by interfacial membranes consisting of soy protein isolate (SPI). This polymer was chosen because it is an abundant natural polymer. There are several previous studies that have addressed the potential for soy protein to be applied to the encapsulating wall material..[7–10]Spray drying is the most common procedure for microencapsulation in the food industry.[11] This process is cost effective, flexible and produces particles of good quality. Many researchers have used the spray drying process to encapsulate oils and flavours such as sunflower oil,[12] avocado oil,[13] coffee oil[11] and lycopene.[14] Optimization has been used in food engineering for the efficient operation of processing systems and unit processes yielding a highly acceptable product. Response surface methodology (RSM) is the process, which combines mathematics with statistics. In multifactor experiments, RSM can be used to examine comprehensively various parameters with minimum experimental times and determine the most relevant factors and their influence ranges, as well as interactions among the factors. This experimental strategy has been widely used in the development of food processes.[15,16] Although previous studies on microencapsulation of food ingredients into the polymeric matrix by spray drying can be found in the literature, none of them reports on the microencapsulation of vanillin. This is an interesting issue that deserves to be studied, as it can represent a promising alternative for the food, ingredient and drug industries. The main objectives of this study were: (i) to investigate the main effects of process variables on the product quality during spray drying of microencapsulate vanillin; (ii) characterize the microcapsules formed; and (iii) to determine optimum process conditions for microencapsulate of vanillin.

Materials and methods

SPI of food grade was purchased from Golhar Co (Mashhad, Iran). Maltodextrin (with a DE = 15–20) was purchased from A.h.m.s (China). Sunflower oil was purchased from a local supermarket and used without further purification. Analytical grade vanillin, hydrochloric acid and sodium hydroxide were purchased from Sigma Chemical Company. Distilled and de-ionized water was used for the preparation of all solutions.

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Emulsions preparation

The stock solution of SPI was prepared by dispersing SPI powder in deionized water and stirred for 5 min then the solution exposed to ultrasonic 500 W (Schaper model Unique USC 25 kHz) for 5 min at 50 °C and then centrifuged at 5000 *g* for 10 min.[17] These solutions were stored at room temperature for 24 h to ensure complete dissolution of these materials. Emulsions were prepared by homogenizing 5 wt% sunflower oil (containing 0.1–0.4 g of vanillin) with 95 wt% aqueous emulsifier solution (1% w/v SPI) in a high-speed blender (2 min at 8944 g) (UltraTurrax T-25, IKA Instruments, Germany S25N-18G-ST) followed by sonication for 2 min at a frequency of 20 kHz, amplitude of 60% (VCX 750; Sonics & Materials, Inc., USA). This emulsion adjusted back to pH 3.5 using 1 M HCl. Prior to spray drying, maltodextrin was added directly to the emulsions at concentrations ranging from 5 to 15 wt %.

Moisture content

The moisture content of the powder was measured by oven drying at 105 °C for 16 h.[9]

Encapsulation efficiency

The method described in Rodríguez *et al.* (2013) to calculate encapsulation efficiency was adapted: approximately 0.1 g of the sample was dissolved in 10 ml of ethanol in a sealed vial. After mixing, the tubes remained at rest in the dark for about 2 h for decantation of the encapsulation material. Absorption was read in a spectrophotometer (Shimadzu UV-160A) in the 231-wavelength nm and using a previously elaborated standard curve, it was possible to calculate the concentration of vanillin present in the microcapsules. The encapsulation efficiency was calculated as the quantity of vanillin present in the capsules compared with the vanillin initially used to produce them.[18]

Particle size distribution

The particle size distribution was measured using a laser light diffraction instrument (SALD-2101; Shimadzu, Japan). A small powder sample was dispersed in 99.5% acetone and the particle distribution was monitored during five successive readings. The particle size was expressed as the mean volumetric size d_{43} (De Brouckere mean diameter), which is the mean diameter of a sphere with the same volume, and is generally used to characterize a particle.

Particle morphology

The particle was evaluated by SEM, and for such, the samples were mounted on the specimen holder with a double-sided adhesive tape and vacuum coated with gold. The observations were made using a scanning electron microscope (Leo 1450VP) and a voltage of 20 kV.

Experimental design and statistical analysis

The spray drying process was performed using a laboratory-scale Mini Spray Dryer (Two-Flow nozzle, Counter-current; Soroush, Iran). In the statistical design of the experiment, a response surface methodology (RSM) was used to estimate the main effects of the process variables on moisture content, particle size and encapsulation efficiency in microencapsulation. A Box-Behnken design was used with a maltodextrin concentration (5–15%, wt), vanillin concentration (0.1–0.4%, wt) and inlet temperature (180–200 °C) being the independent process variables (Table 1). This generated 17 experiments with five replications at the centre point. The RSM was applied to the experimental data using a commercial statistical package, Design-Expert version6.02 (Stat ease Inc., Minneapolis, USA). The following polynomial model was fitted to the data:

$$Y = b_0 + b_1X_1 + b_2X_2 + b_3X_3 + b_{11}X_{12} + b_{22}X_{22} + b_{33}X_{33} + b_{12}X_1X_2 + b_{13}X_1X_3 + b_{23}X_2X_3$$

where b_n are constant regression coefficients; Y is the response (i.e. moisture content, particle size and encapsulation efficiency); X_1, X_2, and X_3 are inlet temperature, maltodextrin concentration and vanillin concentration, respectively.

Mathematical models were evaluated for each response by means of multiple regression analysis. The modelling was started with a quadratic model including linear, squared and interaction terms. Significant terms in the model for each response were found by analysis of variance (ANOVA), and significance was judged by the *F*-statistic calculated from the data.[15,16,19] After model fitting, residual analyses including the examination of diagnostic plots and calculation of case statistics were conducted to validate assumptions used in ANOVA. The software was used to fit response surfaces and optimize the drying process.

Results and discussion

Multiple linear regression analysis of the experimental data yielded second-order polynomial models for predicting moisture content, particle size and encapsulation efficiency. ANOVA was conducted to determine significant effects of process variables on each response and to fit second-order polynomial models to experimental data. Regression equation coefficients of the proposed models and statistical significance of all main effects calculated for each response were obtained, and no significant effects ($P > 0.05$) were stepped down from models without damaging the model hierarchy (Table 2). ANOVA also showed that the lack of fit was not significant for all response surface models at the 95% confidence level. To visualize the combined effects of two factors on any response, the response surface and contour plots were generated for each of the fitted models as the function of two independent variables, while keeping the other variable at the central value. Three different response surface plots (Figures 1–3) were illustrated by maintaining one of the factors constant for each figure. These figures were typical examples plotted for the centre points of the constant factor. The effects of variables on the responses were discussed by evaluation of these plots. Model adequacy checking may be carried out stepping down the effects that are not significant ($P > 0.05$) and predicted R^2.

Table 1. Range of different variables for the spray drying process in the coded and uncoded form

Coded values	Uncoded values		
	Inlet temperature	Vanillin concentration (%wt)	Maltodextrin concentration (%wt)
−1	180	0.1	5
0	190	0.25	10
1	200	0.4	15

Table 2. ANOVA evaluation of the response variable and coefficient of the prediction models

		Moisture content			Encapsulation efficiency			Particle size	
Source	DF	Coefficient	*P*-value	DF	Coefficient	*P*-value	DF	Coefficient	*P*-value
Model	1	2.05	<0.001	3	0.33	0.0015	4	7.49	0.0034
X_1	1	−1.1	<0.001	1	−0.1	0.0016	1	1.6	0.0029
X_2	1		0.19	1			1	0.96	0.045
X_3	1		0.11		0.067	0.0219	1		
X_1^2				1	0.081	0.406			
X_2^2							1	1.68	0.0142
Lack of fit	11		0.14	9		0.074			0.1
R^2			0.84			0.88			0.92
Adj-R^2			0.81			0.81			0.84

DF, Degrees of freedom (statistics).

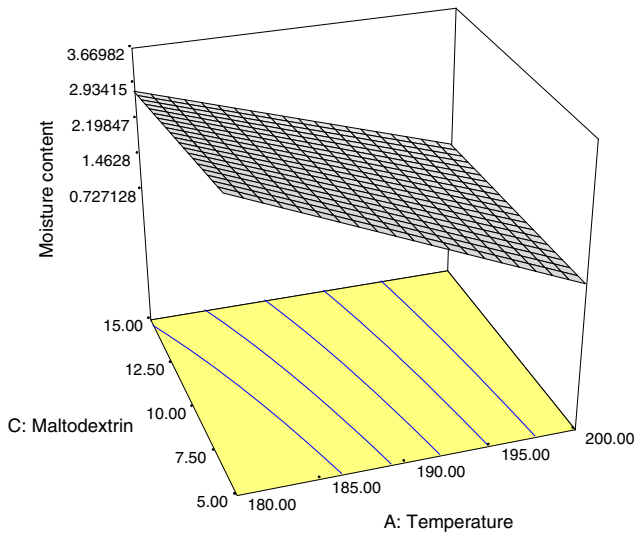

Figure 1. Response surface and contour plots for moisture content

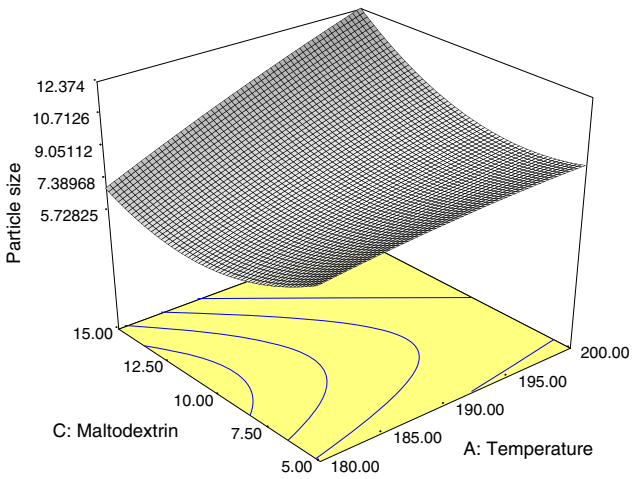

Figure 2. Response surface and contour plots for particle size

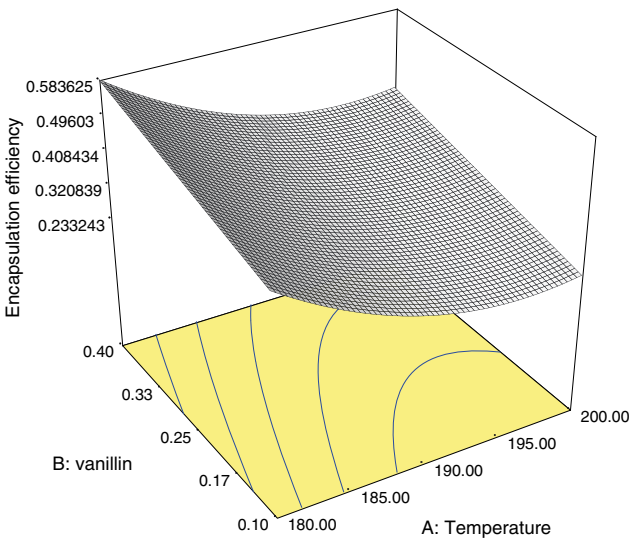

Figure 3. Response surface and contour plots for encapsulation efficiency

Moisture content

Table 2 indicated that only the linear effect of temperature on the moisture content was significant. The magnitude of the coefficient in Table 2 indicates the linear effects negative contribution of temperature on moisture content. Figure 1 indicates a decrease in moisture content with an increase in temperature at process duration because the diffusion is faster. Other researchers showed that increasing the inlet air temperature increased the drying rate and, hence, the moisture content of the powder was reduced.[11,20–22]

Particle size

One of the most important characteristics of powder or particulate system is the distribution of particles.[20] Figures 4 and 5 show the particle size distribution of the microcapsules produced with different maltodextrin concentrations (at 200 °C) and at different inlet air temperatures (with 15% maltodextrin). Most showed a unimodal distribution, indicating good powder homogeneity, with one peak representing a predominant size. The powders exhibited a very large size range, with diameters ranging from 0.7 to 128 μm, which is typical of particles produced by spray drying.[20]

The effects of independent variables on particle size as represented in Figure 2. The increase in the level of temperature and maltodextrin concentration had an increased particle size ($d_{4,3}$) response. The magnitude of the coefficient in Table 2 indicates that

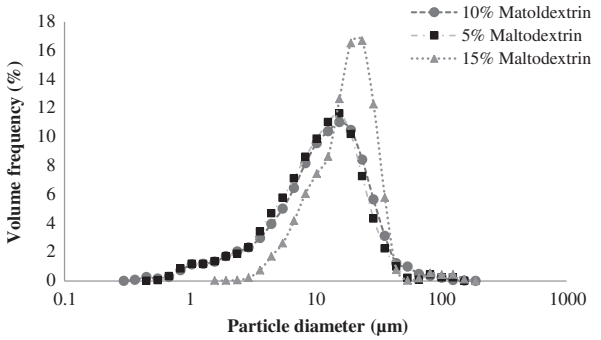

Figure 4. Particle size distributions of the microcapsules produced with different maltodextrin concentration at 200 °C

the linear effect of maltodextrin concentration and temperature show positive effects on particle size while the linear effect of vanillin concentration has not effect on particle size. The quadratic terms of maltodextrin concentration have a positive effect on particle size. Figure 2 shows that at a higher temperature, increasing the concentration of maltodextrin makes the particle size larger. It is for this reason that increasing total solids leads to an increase in emulsion viscosity, reducing the circulation movements inside the droplets and thus, resulting in a rapid skin formation also, the increase in temperature leaded to increased particle size. These factors caused by the formation of a hard crust that does not allow particle shrinkage during spray drying, thus increasing its size.[20] Yang *et al.* (2014) observed an average particle size of 7.35 μm for microcapsules of vanilla oil obtained by complex coacervation using chitosan and Arabic gum as the encapsulating agents.[23]

Encapsulation efficiency

The effects of independent variables on the encapsulation efficiency are represented in Figure 3. The increase in the level of vanillin concentration has an increased encapsulation efficiency response, while the increase in the level of temperature has a decreased encapsulation efficiency response; also, maltodextrin has no effect on the encapsulation efficiency response. The magnitude of the coefficient in Table 2 indicates that the linear effect of vanillin concentration shows positive effects on encapsulation efficiency while the linear effect of temperature shows negative effects on encapsulation efficiency. The decrease in the encapsulation efficiency with the increase in temperature could be related to the fact that higher inlet air temperatures affect the balance between the water evaporation rate and film formation, leading to a breakdown of the crust. In addition, the inlet air temperature increase can cause excessive bubble growth and surface imperfections that cause heat damage to the product, which increases volatile losses during spray drying.[4,20] The non-linearity of Figure 3 shows that quadratic terms of temperature have a positive effect on encapsulation efficiency.

The optimum condition for spray drying of microcapsules of vanillin was determined to obtain maximum encapsulation efficiency and minimum moisture content and particle size. Second-order polynomial models obtained in this study were utilized for each response in order to determine the specified optimum conditions. These regression models are valid only in the selected experimental domain. So, optimization criteria were selected based on different parameters including economical and product-quality-related attributes.[19] By applying the desirability function method, the solution was obtained for the optimum covering the criteria. At this point, 184 °C for inlet temperature, 8.5% of the maltodextrin concentration and 0.36% of the vanillin concentration.

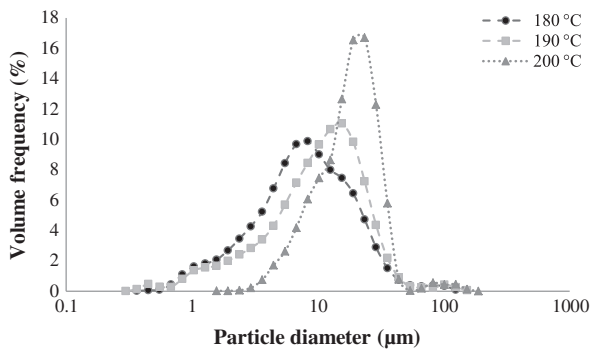

Figure 5. Particle size distributions of the microcapsules produced with 15 % maltodextrin concentration at different inlet air temperatures

Validation of optimal conditions

Analysis of vanillin microencapsulates was carried out at optimal conditions to verify the model. Table 3 shows the analytical characterization of vanillin microencapsulates obtained according to the optimized response. The results that showed a high-performance model predicted the responses.

Morphology of particle

Figure 6 shows the SEM microphotographs of the powders produced at the selected optimal conditions (184 °C min for inlet

Table 3. Powder analysis of vanillin microencapsulates under conditions of the optimal model point

Powder analysis					
Moisture content (g/100 g powder)		Particle size (μ)		Encapsulation efficiency (%)	
Predicted value	Observed value	Predicted value	Observed value	Predicted value	Observed value
3.153	2.171	6.95	7.19	58.3	51.9

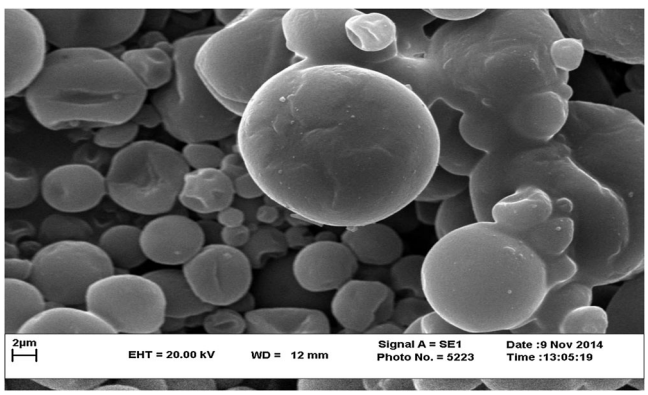

Figure 6. Morphology of vanillin microencapsulates under conditions of optimal model point

temperature, 8.5% for the maltodextrin concentration and 0.36% for the vanillin concentration). Particles showed a spherical shape with no apparent cracks or fissures, which promotes better protection and retention of the core material.

Conclusion

Microencapsulate of vanillin was produced by spray drying using SPI and maltodextrin as a wall material. RSM was used to determine the optimum processing conditions that yield the maximum encapsulation efficiency and the minimum moisture content and particle size during drying of vanillin microencapsulate. The second-order polynomial models for all the response variables were found to be statistically significant. The optimal conditions for maximum encapsulation efficiency and minimum moisture content and particle size, were 184 °C with 8.5% maltodextrin concentration and 0.36% vanillin concentration in order to obtain a moisture content (3.153 g/100 g powder), particle size (6.95 μ) and encapsulation efficiency (58.3%).

References

1. V. T. Karathanos, I. Mourtzinos, K. Yannakopoulou, N. K. Andrikopoulos. *Food Chem.* **2007**, *101*, 652.
2. D. Havkin-Frenkel, F. C. Belanger. *Handbook of Vanilla Science and Technology*. Wiley Online Library: Blackwell Publishing Ltd, UK, **2011**. http://onlinelibrary.wiley.com/book/10.1002/9781444329353
3. D. Stojakovic, B. Bugarski, N. Rajic. *J. Food Eng.* **2012**, *109*, 640.
4. H. C. Carneiro, R. V. Tonon, C. R. Grosso, M. D. Hubinger. *J. Food Eng.* **2013**, *115*, 443.
5. Y. Serfert, C. Lamprecht, C.-P. Tan, J. Keppler, E. Appel, F. Rossier-Miranda, K. Schroen, R. Boom, S. Gorb, C. Selhuber-Unkel. *J. Food Eng.* **2014**, *143*, 53.
6. L. Castan, G. del Toro, A. A. Fernadez, M. Gonzlez, E. Ortz, D. Lobo. *J. Med. Food* **2013**, *16*, 551.
7. A. Nesterenko, I. Alric, F. o. Silvestre, V. Durrieu *Food Res. Int.* **2012**, *48*, 387.
8. A. Nesterenko, I. Alric, F. Silvestre, V. Durrieu. *Food Hydrocoll.* **2014**, *38*, 172.
9. C.-H. Tang, X.-R. Li. *Food Res. Int.* **2013**, *52*, 419.
10. Z. Teng, Y. Luo, Q. Wang. *J. Agric. Food Chem.* **2012**, *60*, 2712.
11. E. Frascareli, V. Silva, R. Tonon, M. Hubinger. *Food and bioproducts processing* **2012**, *90*, 413.
12. J.-H. Ahn, Y.-P. Kim, Y.-M. Lee, E.-M. Seo, K.-W. Lee, H.-S. Kim. *Food Chem.* **2008**, *107*, 98.
13. E. Bae, S. Lee. *J. Microencapsul.* **2008**, *25*, 549.
14. G. A. Rocha, C. S. Favaro-Trindade, C. R. F. Grosso. *Food and bioproducts processing* **2012**, *90*, 37.
15. M. Noshad, M. Mohebbi, F. Shahidi, S. A. Mortazavi. *Food Tech. Biotechnol.* **2012**, *5*, 2098.
16. O. Corzo, N. Bracho, A. V‹squez, A. Pereira. *J. Food Eng.* **2008**, *85*, 372.
17. G.-Q. Huang, Y.-T. Sun, J.-X. Xiao, J. Yang. *Food Chem.* **2012**, *135*, 534.
18. S. D. Rodriguez, T. F. Wilderjans, N. Sosa, D. L. Bernik. *J. Food Res.* **2013**, *2*, 36.
19. E. Eren, F. Kaymak-Ertekin. *J. Food Eng.* **2007**, *79*, 344.
20. S. M. Jafari, E. Assadpoor, Y. He, B. Bhandari. *Drying Technol.* **2008**, *26*, 816.
21. C. C. Ferrari, S. P. M. Germer, J. de Aguirre. *Drying Technol.* **2012**, *30*, 154.
22. P. Roccia, M. L. Martinez, J. M. Llabot, P. D. Ribotta. *Powder Technol.* **2014**, *254*, 307.
23. Z. Yang, Z. Peng, J. Li, S. Li, L. Kong, P. Li, Q. Wang. *Food Chem.* **2014**, *145*, 272.