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# EFFECTS OF THE PROCESS VARIABLES OF MICROENCAPSULATION SESAME OIL (*Sesamum indica* L.) BY SPRAY DRYING

# EFECTOS DE LAS VARIABLES DE PROCESO EN LA MICROENCAPSULACIÓN DEL ACEITE DE AJONJOLÍ (*Sesamum indica* L.) MEDIANTE SECADO POR ASPERSIÓN

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**Abstract**

The aim of this study was to investigate the effects of the process variables of microencapsulation sesame oil (SO) by spray drying to generate the desired powder quality with the highest efficiency encapsulation and maximum linoleic acid content microencapsulated using a full factorial design of experiments. Thirty-two tests were made, and five replicates were conducted on the central points. Independent variables were volumetric dispersed phase ($\phi_{O/W}$) (0.05, 0.10 and 0.15), wall material to core ratios (Wa:Co) (1:1, 2:1 and 3:1) and drying air inlet temperature (Ti) (120, 140 and160 °C). Surface oil (SOM%), encapsulation efficiency (EE%), linoleic acid content (LAC%), and moisture content (MC%) were analyzed as responses. Under maximum process conditions Wa:Co=2.59:1, $\phi_{O/W}$ =0.05 and Ti=154.04 °C, the response variables including, EE and LAC were predicted as 88.20% and 50.02% respectively. It was concluded that these microcapsules containing high content of linoleic acid can be used as functional food.

*Keywords*: sesame oil, spray drying, encapsulation efficiency, linoleic acid, microencapsulation.

**Resumen**

El objetivo de este trabajo fue investigar los efectos de las variables de proceso en la microencapsulación del aceite de ajonjolí (SO) mediante secado por aspersión para generar un polvo de calidad, con la eficiencia de encapsulación más alta y máximo contenido de ácido linoleico microencapsulado utilizando un diseño de experimentos factorial. Se realizaron 32 experimentos con 5 réplicas en el punto central. Las variables independientes fueron la fracción volumétrica de la fase dispersa ($\phi_{O/W}$) (0.05, 0.10 y 0.15), la relación de material de pared respecto al material encapsulado) (1:1, 2:1 y 3:1) y la temperatura del aire a la entrada del secador (Ti) (120, 140 y160 °C). Las variables de respuesta que se analizaron fueron los porcentajes de aceite superficial (SOM%), eficiencia de encapsulación (EE%), contenido de humedad en las microcápsulas (MC%) y contenido de ácido linoleico microencapsulado (LAC%). Bajo máximas condiciones de proceso Wa:Co=2.59:1, $\phi_{O/W}$ =0.05 y Ti=154.04 °C, los valores de eficiencia de encapsulación y contenido de ácido linoleico microencapsulado predichos fueron de 88.20% y 50.02%, respectivamente. Se concluye que estas microcápsulas contienen un alto contenido de ácido linoleico y que se puede utilizar para formular alimentos funcionales.

*Palabras clave*: aceite de ajonjolí, secado por aspersión, eficiencia de encapsulación, ácido linoleico, microencapsulación.

## 1 Introduction

Sesame (*Sesamum indicum* L.) is the most ancient oilseed crop known to humans, has been cultivated in Asia and Africa for 2000 years ago. Sesame oil has been used as a natural ingredient in salads and also as a seasoning oil to prepare foods. The main compounds of the sesame oil are unsaturated fatty acids (UFA), ~47% of linoleic acid and ~37% of oleic acid (Corso *et al*., 2010; Lee *et al*., 2012). Both

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essential fatty acids are important in the human feed, because they offer health benefits. In the last years has been reported that the consumption of sesame oil influences in blood lipid profiles, increasing anti-inflammatory function, and exhibiting antimutagenic activity (Xu-Yan *et al*., 2012). Unsaturated essential fatty acids in sesame oil are chemically unstable in presence of oxygen, light, moisture and heat. Stability of sesame oil can be enhanced through microencapsulation process. Microencapsulation by spray-drying is widely used in the food industry for the preparation of dry stable additives such as oils and flavours. Microencapsulation process involves two principal stages. The first stage involves the preparation of an emulsion, which depends on certain parameters such as the volumetric dispersed phase, the total solids content, and the ratio of wall material to core material about which influence the characteristics and properties functional of microcapsules. The second stage involves the atomization of the emulsion in a spray dryer inside of chamber, where a transfer of mass-energy exchange is carried out in order to obtain microcapsules. The water of the emulsion that was fed inside of chamber is instantly vaporized, allowing that the oil be trapped within in an encapsulate material (Gharsalloui *et al*., 2007). Optimisation of the spray drying for encapsulated products has been successfully reported by using response surface methodology (RSM) (Bringas-Lantigua *et al*., 2011; Kha *et al*., 2014). However, there are few studies in the literature involving jointly in design of experiments the parameters in the formulation of the emulsion and variable process conditions in the spray-drying stage, practically all studies focuses only spray drying and not microencapsulation process (Tonon *et al*., 2011; Lee *et al*., 2013; Balasubramani *et al*., 2013). Statistical analysis used in testing multiple process factors and their interactive effects (Corona-González *et al*., 2013; Roccia *et al*., 2014; Rodríguez-Bernal *et al*., 2015; Flores-Martínez *et al*., 2016) can help in investigating the interactive effect of process variables and in building mathematical model that accurately describes the overall process such as microencapsulation process (Lee *et al*., 2013). Therefore, the aim of this research was to investigate the effects of the process variables of microencapsulation sesame oil by spray drying to generate the desired powder quality with highest efficiency encapsulation and maximum linoleic acid content microencapsulated studying the influence of the wall material to core ratios, volumetric dispersed phase and drying air inlet temperatures. Encapsulation efficiency, surface oil, linoleic acid content and moisture content were analyzed as responses. In addition the microstructure of the optimized SO microcapsules was analyzed. In order to contribute to maximize the technological microencapsulation process for food dried, guaranteeing the quality and longer shelf-life of the powder product, it is hoped that the microcapsules can be potentially incorporated in dry form enhance the functionality of foods (cake mixes, snacks, baby foods, etc.).

# 2 Materials and methods

## *2.1 Materials and reagents*

The sesame seeds were purchased from regional supermarket in Toluca City (Toluca, State of Mexico, Mexico). Mesquite gum (MG) hand collected in the form of tear drops from *Prosopis laevigata* trees in the Mexican State of San Luis Potosi and purified as indicated by Vernon-Carter *et al*. (1996) and Maltodextrin DE-10 was purchased from Industria Ragar, S.A. de C.V. (Mexico City, Mexico). The organic chemicals used in the analyses were analytical and chromatographic grades and were purchased from Sigma Aldrich (Toluca, State of Mexico, Mexico). All the water used in the experiments was deionized.

## *2.2 Extraction of sesame oil*

A Tamer hydraulic press (Model PT-20, Shanghai, China) fitted with a 40 cm long and 10 cm diameter plunger was used for cold pressing the sesame seeds for obtaining the sesame oil. Maximum pressure applied by the piston was $8.8\times10^8$ N/m$^2$ to the piston, at room temperature (~20 ºC). Trace amounts of seed were removed from sesame oil using a cloth filter, and filtered sesame oil was stored in amber bottles at a temperature of ~5 ºC until required.

## *2.3 Experimental design*

A statistical analysis was applied using a quadratic design with 3 factors and 3 levels that consisted of 27 points and 5 replicates on central points ($n$ = 32) to maximize the process variables in the microencapsulation of sesame oil (SO) by spray drying and was conducted in a random order. The following parameters were selected as independent variables: ($X_1$) volumetric dispersed phase ($\phi_{O/W}$), ($X_2$) wall material to core ratio (Wa:Co), and ($X_3$) drying air inlet temperature ($T_i$).

Table 1. Levels of the operating independent variables

| Independent variables | | Levels of the operating | | |
|---|---|---|---|---|
| | | -1 | 0 | 1 |
| $X_1$ | $\phi_{O/W}$ (volumetric dispersed phase) | 0.05 | 0.1 | 0.15 |
| $X_2$ | Wa:Co (wall material to core material) | 1:1 | 2:1 | 3:1 |
| $X_3$ | Temperature (°C) | 120 | 140 | 160 |

Table 1 shows the levels of the operating independent variables. The levels of independent variables were based on preliminary trials. The response variables were encapsulation efficiency (EE, %), surface oil microcapsules (SOM, %), linoleic acid content (LAC, %) and moisture content (MC, %). The experimental design is shown in Table 2. Full factorial design of experiments has previously been successfully applied in a wide range of applications; for example, quality and processes optimization studies in food systems and consumer preference studies (Behboudi-Jobbehdar *et al*., 2013). Although full factorial design approaches are generally regarded as time and cost consuming due to the large number of experiments required, they do offer the best practice approach for process or product optimization where the factor interactions cannot be neglected (Behboudi-Jobbehdar *et al*., 2013). In this work two main criteria were used to optimize the microencapsulation process, achieve the highest efficiency encapsulation and maximum linoleic acid content microencapsulated. The regression models were evaluated for each response variable, and the resulting equations were tested for adequacy and fit by an analysis of variance (ANOVA) (Table 3). The relationship between the independent variables and the response variables was calculated by the second-order polynomial (Eq. (1))

$$Y = \beta_0 + \sum_{i=1}^{k} \beta_i X_i + \sum_{i=1}^{k} \beta_{ii} X_i X_i + \sum_{i=1}^{k} \sum_{j=i+1}^{k} \beta_{ij} X_i X_j \quad (1)$$

where $Y$ is the predicted response, $\beta_0$ is a constant, $\beta_i$ is the linear coefficient, $\beta_{ii}$ is the square coefficient, $\beta_{ij}$ is the cross product coefficient, and $k$ is the number the factors ($k = 3$).

## 2.4 Emulsion preparation

Sesame oil-in-water (O/W) emulsions variations were prepared using a biopolymers blend mesquite gum (MG) with maltodextrin DE10 (MD) (MG66%-MD34% w/w), wall material to core ratios (Wa:Co) of 1:1, 2:1 and 3:1, and volumetric dispersed phase ($\phi_{O/W}$) of 0.05, 0.10 and 0.15.

Table 2. Experimental design and values obtained for the response variables

| Independent variables | | | | Response variables | | | |
|---|---|---|---|---|---|---|---|
| | $X_1$ | $X_2$ | $X_3$ | SOM % | EE % | LAC % | MC % |
| **1** | -1 | -1 | -1 | 23.508 | 76.492 | 49.258 | 5.385 |
| **2** | 1 | -1 | -1 | 14.198 | 85.802 | 50.23 | 4.906 |
| **3** | -1 | 1 | -1 | 17.338 | 82.662 | 49.11 | 3.652 |
| **4** | 1 | 1 | -1 | 16.76 | 83.24 | 48.567 | 5.638 |
| **5** | -1 | -1 | 1 | 11.892 | 88.108 | 47.684 | 4.599 |
| **6** | 1 | -1 | 1 | 16.158 | 83.842 | 50.06 | 4.116 |
| **7** | -1 | 1 | 1 | 13.394 | 86.606 | 50.095 | 3.067 |
| **8** | 1 | 1 | 1 | 29.27 | 70.73 | 50.199 | 5.251 |
| **9** | -1 | -1 | 0 | 16.44 | 83.56 | 48.8 | 4.496 |
| **10** | 1 | 1 | 0 | 20.316 | 79.684 | 49.61 | 4.627 |
| **11** | -1 | 1 | 0 | 14.106 | 85.894 | 49.621 | 2.852 |
| **12** | 1 | -1 | 0 | 13.918 | 86.082 | 50.249 | 3.9 |
| **13** | -1 | 0 | -1 | 18.767 | 81.233 | 49.845 | 4.46 |
| **14** | 1 | 0 | -1 | 13.823 | 86.177 | 50.006 | 5.09 |
| **15** | -1 | 0 | 1 | 9.63 | 90.37 | 49.89 | 3.3 |
| **16** | 1 | 0 | 1 | 19.619 | 80.381 | 50.395 | 4.264 |
| **17** | -1 | 0 | 0 | 13.617 | 86.383 | 50 | 3.452 |
| **18** | 1 | 0 | 0 | 15.461 | 84.539 | 50.399 | 4.09 |
| **19** | 0 | -1 | 0 | 15.712 | 84.288 | 49.708 | 3.076 |
| **20** | 0 | 1 | 0 | 17.744 | 82.256 | 49.712 | 2.665 |
| **21** | 0 | 0 | -1 | 16.828 | 83.172 | 49.469 | 3.614 |
| **22** | 0 | 0 | 1 | 15.836 | 84.164 | 50.007 | 2.9 |
| **23** | 0 | -1 | -1 | 19.386 | 80.614 | 49.991 | 4.067 |
| **24** | 0 | 1 | 1 | 20.426 | 79.574 | 50.06 | 2.847 |
| **25** | 0 | -1 | 1 | 14.558 | 85.442 | 49.086 | 3.31 |
| **26** | 0 | 1 | -1 | 17.582 | 82.418 | 48.627 | 3.676 |
| **27** | 0 | 0 | 0 | 15.072 | 84.928 | 50.112 | 2.655 |
| **28** | 0 | 0 | 0 | 15.09 | 84.91 | 50.017 | 2.66 |
| **29** | 0 | 0 | 0 | 15.098 | 84.902 | 50.042 | 2.663 |
| **30** | 0 | 0 | 0 | 15.069 | 84.931 | 50.123 | 2.635 |
| **31** | 0 | 0 | 0 | 15.079 | 84.93 | 50.065 | 2.662 |
| **32** | 0 | 0 | 0 | 15.085 | 84.906 | 50.083 | 2.645 |

$X_1$, $X_2$, $X_3$: Independent variable of volumetric dispersed phase, wall material to core ratio and drying inlet air temperature. EE, SOM, LAC, MC: Efficiency encapsulation, surface oil on microcapsules, linoleic acid content and moisture content.

The procedure consisted of an initial stage, where the continuous phase was made by dissolving the mixture of biopolymers in water 24 h prior to the preparation of the emulsion and was added sodium azide to 0.3% based on the total grams of the emulsion with the order to avoid the proliferation of microorganisms; in a second stage the dispersed phase was added dropwise to the continuous phase and emulsified in an Ultra-turrax T50 basic (IKA-WERKE

Works Inc., Wilmington, NC, USA) at a speed of 5200 rpm for 10 minutes, with an ice bath to keep the temperature below 30ºC.

### *2.5 Preparation of microcapsules by spray drying*

The different emulsions variations were spray-dried using in a pilot spray drier Nichols/Niro (Turbo Spray PLA, NY, USA) with a 5 mm diameter nozzle atomizer. The emulsions were fed into the main spray chamber through a peristaltic pump varying the feed flow rate from about 15 to 25 mL/min with an atomization pressure of 4 bar. The drying air inlet temperature was controlled by settings of the air heater of the equipment and were of 120, 140 and 160 °C ± 5 ºC and drying air outlet temperature of 75, 80, 85 ± 5 ºC respectively.

The microcapsules were recovered from the collecting chamber and were stored in a desiccator containing silica gel to prevent moisture adsorption, then vacuum sealed in high density polyethylene (HDPE) plastic bags and stored at 4 °C until analysis (within 24 h).

### *2.6 Surface oil of microcapsules*

The non-encapsulated oil was determined according to the method described by Calvo *et al*. (2012) with some modifications. Briefly, 5g of microcapsules were dispersed in 10 mL of hexane for 10 s (taking care that the particle is not destroyed). The solvent was filtered through Whatman filter paper no. 1, after which the residue was evaporated using a Büchi rotary evaporator R-100 (BÜCHI Labortechnik, AG) under vacuum at 80 °C, subsequently non-encapsulated oil was weighed. The oil was calculated gravimetrically (Jiménez *et al*., 2006).

### *2.7 Total sesame oil in microcapsules*

The total oil was determined according to the method described by Rodea-González *et al*. (2012) with slight modifications. Briefly, 5 g of microcapsules were weighed to which they extract the oil using equipment Soxhlet fat extraction using hexane as solvent extractor for 6 h at room temperature. The solvent residue was evaporating with a vacuum evaporation at 80 ºC. The oil obtained after evaporation was weighed and taken as the total oil content in the microcapsules and was calculated gravimetrically (Jiménez *et al*., 2006).

### *2.8 Encapsulation efficiency*

The percentage of encapsulation efficiency (EE) was determined with the following relationship (Rodea-González *et al*., 2012):

$$\%EE = \frac{(O_{total} - O_{surface})}{O_{total}} \times 100 \qquad (2)$$

where total oil ($O_{total}$) is the sesame oil internal and surface content of microcapsules, while surface oil ($O_{surface}$) is the sesame oil corresponded to the un-encapsulated oil content found at the surface of the particles.

## 3 Determination of acid linoleic content by gas chromatography

The oil without encapsulate and microencapsulate were derivatizated to determine the fatty acids composition. Briefly, 200 $\mu$L of the oil were mixed with 1 mL of 0.2 N HCl-methanol and then incubated at 60 ºC during 4 h, then 0.2 mL of deionized water and 2 mL of hexane were added. After vortexing, the methyl esters were extracted in the hexane layer and used for GC analysis, according to Miranda *et al*. (2013). One $\mu$L of the hexane layer was injected into a Varian 3800 GC (Palo Alto, CA) fitted with an Agilent HP-Innowax polar capillary column (30 m × 0.32 mm × 0.25 $\mu$m). Injector (CP-8410) and FID temperatures were both set at 250 °C. The oven temperature was kept at 50 °C for 2 min, then reached 220 °C at a rate of 30 °C/min and held at this temperature for 25 min. After that, temperature reached 255 °C and it was kept at this temperature for 7 min. Fatty acids were identified by comparing their retention times with those of the Supelco 37 FAME Mix standard.

### *3.1 Moisture content of the microcapsules*

Triplicate samples of microcapsules (~5 g) were weighed and then dried in a ventilated oven at 105 °C for 3 h. The samples were removed from the oven, cooled in a desiccator and weighed. The drying and weighing processes were repeated until constant weigh were obtained.

### *3.2 Scanning electron microscopy (SEM)*

The microcapsules were mounted on carbon sample holders using double-side sticky tape and were observed using a JEOL JMS 7600F scanning electron microscope (Akishima, Japan) with the GB-H mode

at 1 kV accelerating voltage. Micrographs at different magnifications were presented. Samples were not metalized since the microscopy equipment operates under ultra-vacuum conditions (Guadarrama-Lezama *et al*., 2014).

### *3.3 Statistical analysis*

The statistical analysis was performed using Design Expert version 6.0.10 software (Stat-Ease, Inc., Minneapolis, USA). The adequacy of the models was determined by evaluating the Fisher's test value (F-value), coefficient of determination ($R^2$), adjusted-$R^2$ and coefficient of variation (CV) obtained from the analysis of variance (ANOVA). The test of statistical significance was based on the total error criteria with a confidence level of 95%. The regression coefficients were used to make statistical calculations to generate response surface plots from the regression models. All analytical measurements were carried out in triplicate.

The maximum conditions for the micro-encapsulation of sesame oil were determined by analyzing the results of variables that were significantly affected by the experimental conditions and were obtained by Derringer's desired function methodology applying Response Desirability Profiling using Design Expert version 6.0.10 software (Stat-Ease, Inc., Minneapolis, USA) (Derringer and Suich, 1980). The method finds the desired goals for each variable and response. All the independent variables were kept within range while the responses were either maximized or minimized. The numerical optimization finds a point that maximizes the desirability function. In the present study, desirability functions were developed for the criteria of maximum efficiency encapsulation, maximum linoleic acid content microencapsulated, minimum surface oil on the microcapsules, and acceptable moisture content.

## 4 Results and discussion

### *4.1 Fitting of second order polynomial equations and statistical analysis*

The empirical relationship between the experimental results obtained on the basis of experimental design model and the independent variables were expressed by a second-order polynomial equation with interaction terms.

$$SOM = 14.96 + 1.16X_1 + 1.18X_2 + 1.90X_2^2 + 1.28X_3^2 + 2.42X_1X_2 + 3.75X_1X_3 + 2.16X_2X_3 \quad (3)$$

$$EE = 85.04 - 1.16X_1 - 1.18X_2 - 1.90X_2^2 - 1.28X_3^2 - 2.42X_1X_2 - 3.75X_1X_3 - 2.16X_2X_3 \quad (4)$$

$$LAC = 50.10 + 0.035X_1^2 - 0.52X_2^2 - 0.40X_1X_2 + 0.17X_1X_3 + 0.52X_2X_3 \quad (5)$$

$$MC = 2.65 + 0.37X_1 - 0.20X_2 - 0.38X_3 + 1.08X_1^2 + 0.25X_2^2 + 0.58X_3^2 + 0.63X_1X_2 \quad (6)$$

The adequacy and fitness of the models were tested by regression analysis of variance (ANOVA) (Tables 3 and 4). The results indicated that the equation adequately represented the actual relationship between the independent variables and responses. ANOVA is a statistical technique that subdivides the total variation in a set of data into component parts associated with specific sources of variation for the purpose of testing hypotheses on the parameters of the model. Analysis of variance followed by Fisher's statistical test (F-test) was applied to evaluate the significance of each variable. The F-value is the ratio of the mean square due to regression to the mean square due to real error and indicates the influence (significance) of each controlled factor on the tested model (Triola, 2014). The ANOVA results for EE%, SOM%, LAC% and MC% showed Fisher F-value of 189.77, 189.78, 83.95 and 382.68 respectively, which implies that the model is significant and higher. The large value of F indicates that most of the variation in the response can be explained by the regression equation. Coefficient of determination ($R^2$) and adjusted-$R^2$ were also calculated to check the adequacy and fitness of the model. A high $R^2$ coefficient ensures a satisfactory adjustment of the quadratic model to the experimental data. The values of $R^2$ were calculated to be 0.990, 0.990, 0.970 and 0.990 for EE%, SOM%, LAC% and MC% respectively, which imply that 95% of experimental data was compatible. The use of an adjusted-$R^2$ is to evaluate the model adequacy and fitness. The adjusted-$R^2$ value corrects the $R^2$ value for the sample size and for the number of terms in the model. The value of adjusted-$R^2$ (0.98 for EE%, 0.98 for SOM%, 0.96 for LAC% and 0.99 for MC%) is also high and advocates a high correlation between the observed and the predicted values. The coefficient of variation (CV) indicates the relative dispersion of the experimental points from the predictions of the second order polynomial models. As a general rule,

the CV should not be greater than 10% and a high CV indicates that variation in the mean value is high and does not satisfactorily develop an adequate response model. The very low values (0.0057, 0.0291, 0.0024 and 0.0268) of CV clearly representing a very high degree of precision and a good reliability of conducted experiments. Adequate precision measures the signal to noise ratio and compares the range of the predicted values at the design points to the average prediction error. The ratio greater than 4 is desirable and indicates adequate model discrimination (Beg *et al*., 2003). In this work the ratio is found to be > 65, which indicates an adequate signal. Therefore, quadratic model can be used to navigate in the design space.

## *4.2 Adequacy of the models*

Generally, it is important to confirm the fitted model to make sure that it gives a sufficient approximation to the actual values. Unless the model shows a satisfactory fit, proceeding with an investigation and optimization of the fitted response surface likely gives poor or misleading results (Murugesan *et al*., 2007). Diagnostic plots such as the predicted *versus* experimental values (Fig. 1) help us to judge the model satisfactoriness and exhibit the relationship between predicted and experimental values. In this figure, each of the observed values was compared to the predicted value calculated from the model. The data points on this plot lie reasonably close to the straight line and indicate an adequate agreement between the real data and the data obtained from the model. The result suggests that the model used in this research were able to identify operating conditions for microencapsulation sesame oil by spray drying.

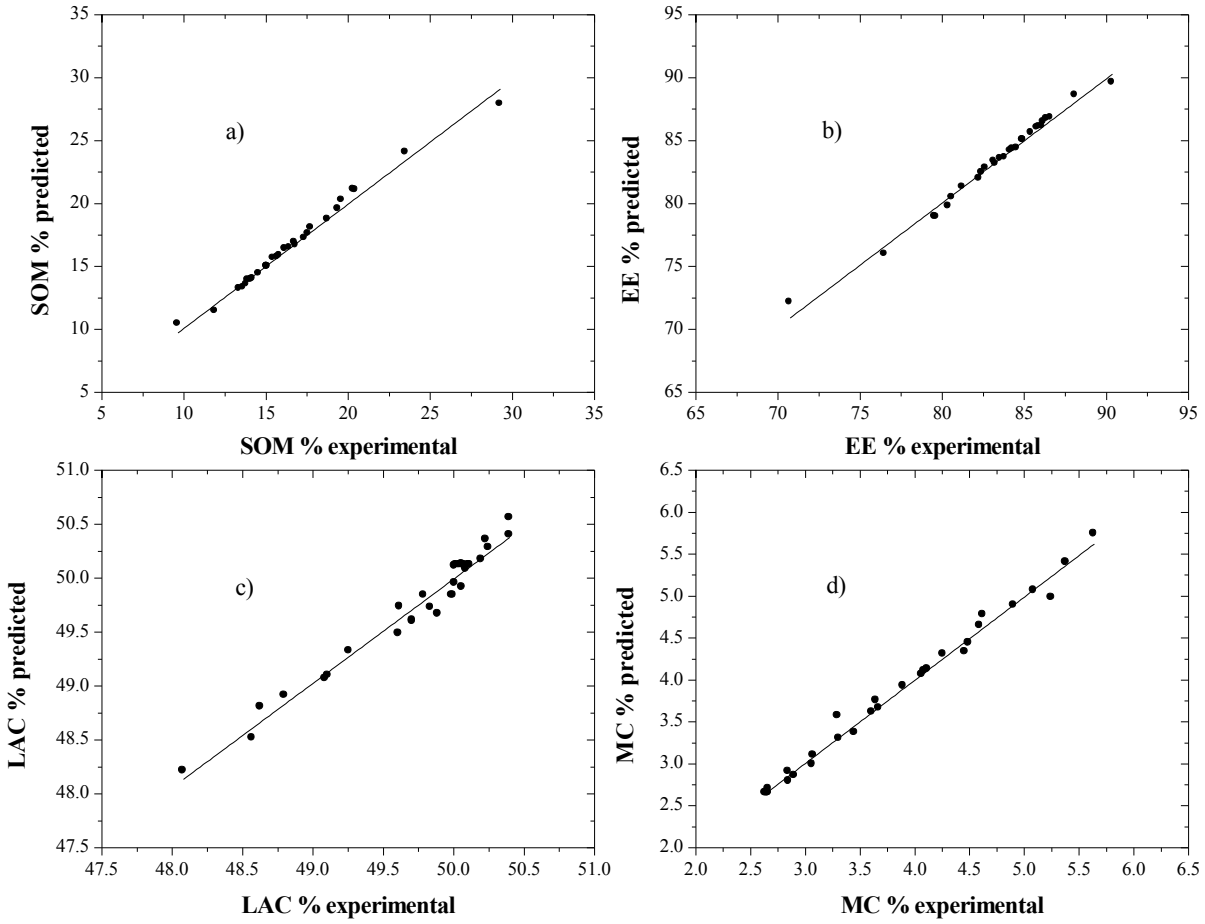


Fig. 1. Diagnostic plots of the relationship between predicted and experimental values: (a) SOM %, (b) EE %, (c) LAC % and (d) MC %.

Table 3. Regression analysis of variance and fitness of the models

| Source | Sum of squares | DF | Mean square | F value | p-Value |
|---|---|---|---|---|---|
| **SOM%** | | | | | |
| Model | 393.32 | 9 | 43.7 | 189.78 | <0.0001 |
| $X_1$ | 24.1 | 1 | 24.1 | 104.68 | <0.0001 |
| $X_2$ | 24.87 | 1 | 24.87 | 108.02 | <0.0001 |
| $X_3$ | 3.05 | 1 | 3.05 | 13.25 | 0.0014 |
| $X_1X_2$ | 70.42 | 1 | 70.42 | 305.82 | <0.0001 |
| $X_1X_3$ | 168.6 | 1 | 168.6 | 732.17 | <0.0001 |
| $X_2X_3$ | 55.9 | 1 | 55.9 | 242.76 | <0.0001 |
| $X_1^2$ | 70.42 | 1 | 70.42 | 305.82 | 0.0089 |
| $X_2^2$ | 168.6 | 1 | 168.6 | 242.76 | <0.0001 |
| $X_3^2$ | 55.9 | 1 | 55.9 | 242.76 | <0.0001 |
| Residual error | 5.07 | 22 | 0.23 | — | — |
| Lack of fit | 5.07 | 17 | 0.3 | 2180.22 | <0.0001 |
| C.V. % | 2.91 | — | — | — | — |
| Adeq Precision | 65.11 | — | — | — | — |
| | — | — | — | — | — |
| **EE%** | | | | | |
| Model | 393.32 | 9 | 43.7 | 189.78 | <0.0001 |
| $X_1$ | 24.1 | 1 | 24.1 | 104.68 | <0.0001 |
| $X_2$ | 24.87 | 1 | 24.87 | 108.02 | <0.0001 |
| $X_3$ | 3.05 | 1 | 3.05 | 13.25 | 0.0014 |
| $X_1X_2$ | 70.42 | 1 | 70.42 | 305.82 | <0.0001 |
| $X_1X_3$ | 168.6 | 1 | 168.6 | 732.17 | <0.0001 |
| $X_2X_3$ | 55.9 | 1 | 55.9 | 242.76 | <0.0001 |
| $X_1^2$ | 70.42 | 1 | 70.42 | 305.82 | 0.0089 |
| $X_2^2$ | 168.6 | 1 | 168.6 | 242.76 | <0.0001 |
| $X_3^2$ | 55.9 | 1 | 55.9 | 242.76 | <0.0001 |
| Residual error | 5.07 | 22 | 0.23 | — | — |
| Lack of fit | 5.07 | 17 | 0.3 | 2180.22 | <0.0001 |
| C.V. % | 2.91 | — | — | — | — |
| Adeq Precision | 65.11 | — | — | — | — |

### *4.3 Surface oil (SOM, %) and encapsulation efficiency (EE;%)*

The SOM% did not change as a consequence of process variables combination, relatively all experiments showed low values of SO on microcapsule surface, these values varied 9.63 to 29.27% (Table 2). Although statistically was significantly influenced by the interaction between the volumetric dispersed phase ($X_1$) and the drying air inlet temperature ($X_3$), the combination between the volumetric dispersed phase ($X_1$) and wall material to core ratio Wa:Co ($X_2$), and the interaction of the wall material to core ratio Wa:Co ($X_2$) with the drying air inlet temperature ($X_3$) (Eq. 3). On the other hand, $X_2^2$, $X_2$ and $X_1$ have a less important effect than the interactions described above with respect to the percentage of surface oil (Table 3). The 3D surface and 2D contour plots (Fig. 2.) were drawn to determine the optimal levels of the independent variables. The main effect plot shown that when the volumetric dispersed phase ($X_1$) decreased from 0.15 to 0.05 at drying air inlet temperature between 140 and 160 °C with wall material to core ratio Wa:Co ($X_2$) = 2:1, the SOM% decreased ~14.4%. With respect to the empirical equation to determine the percentage of surface oil in the microcapsules, it was determined that the minimum percentage of 9.63% is obtained with the following values of the variables: $X_1$ = -1 (0.05), $X_2$ = 0 (2:1), and $X_3$ = 1 (160 °C). This decrease in SOM is due lower proportion of oil in emulsions and lower load of oil close to the drying surface, there by longening the diffusion path length

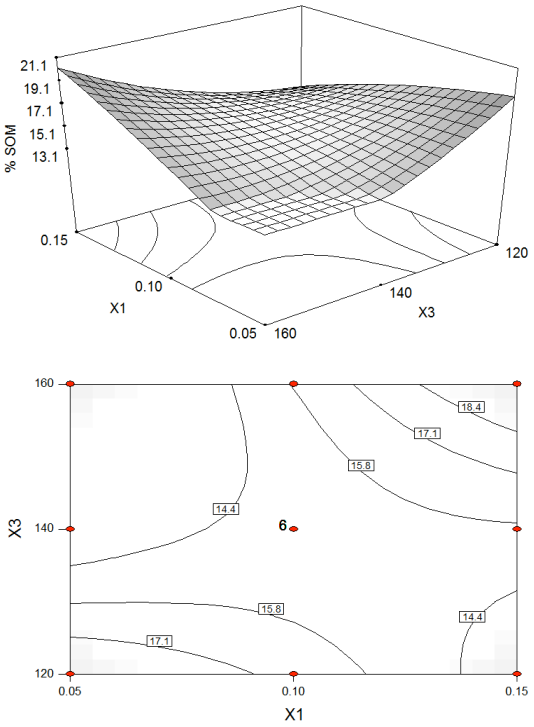


Fig. 2. The (a) 3D response and (b) 2D contour plots of the SOM % affected by volumetric dispersed phase ($X_1$) and drying air inlet temperature ($X_3$).

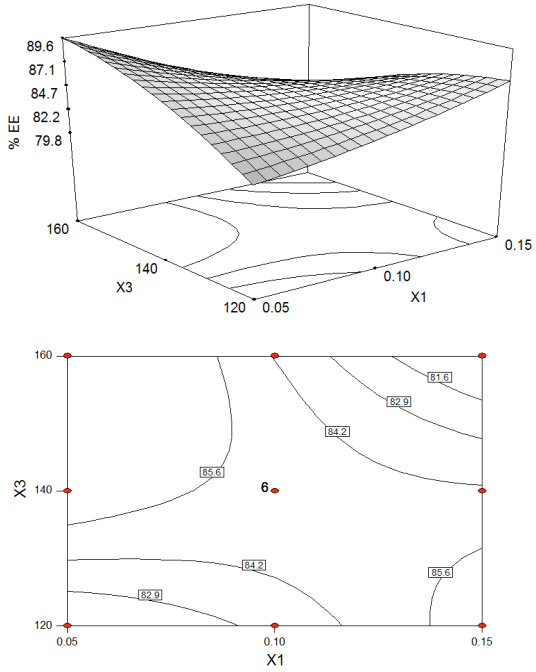


Fig. 3. The (a) 3D response and (b) 2D contour plots of the EE % affected by volumetric dispersed phase ($X_1$) and drying air inlet temperature ($X_3$).

to the air/particle interface and therefore decreasing the amount of surface oil powder. In the temperature range between 140 and 160 °C apparently there was no excessive evaporation, so it did not result in a rupture of the matrix formed by the wall material, reducing the amount of surface oil.

The encapsulation efficiency reflects the amount of oil within of the microcapsules and the degree to which the matrix can prevent diffusion of internal oil through of the wall. According to Table 2, EE% varied from 70.73% to 90.37% which can be considered an adequate level for oil powders. It is important to note that two-factor interactions exerted a significant influence on the EE% of sesame oil ($X_1X_3 > X_1X_2 > X_2X_3$), although the main effect was the combination between $X_1$ and $X_3$. Fig. 3 shows that the interaction between the volumetric dispersed phase ($X_1$) and the drying air inlet temperature (X3), where is noticeable that at $X_1 = 0.05 - 0.1$ and $X_3 = 140$-160 °C at $X_2 = 2:1$ constant, the EE% was observed between 85-87%. Furthermore, the highest value of EE% was obtained at values of $X_1 = 0.10$ and $X_3 = 160$ °C at $X_2 = 1:1$. Lower $X_1$ with higher $X_3$ resulted in higher encapsulation efficiency and implies in shorter time to form a matrix, making difficult the oil diffusion to the drying particle surface. The high amounts of sesame oil retained into microcapsules also are attributed to the nature of the wall material utilized and the amount of wall material in the emulsions which was able to emulsify and consequently retain the oil used. Several researches informed about the good properties of MG and MD for encapsulated oils and oleoresins (Jafari *et al.*, 2008; Sánchez-Sáenz *et al.*, 2011). In this work, MG was proposed as part of wall material due their good emulsifying properties and effective microencapsulation of oils (Rodea-González *et al.*, 2012; Escalona-García *et al.*, 2016). Mesquite gum has been reported to offer emulsifier properties because it is composed by a backbone of residues of (1-3) linked $\beta$-D-galactose, and (1-6) side chains containing L-arabinose, L-rhamnose, $\beta$-D-glucuronate and 4-o-methyll-$\beta$-D-glucuronate, having an small amount of protein (2.7 ± 0.06%) attached to the polysaccharide moiety, which is mainly responsible caused by its excellent emulsifying and film forming capacity (Pérez-Alonso *et al.*, 2008). Maltodextrins consist of $\beta$-D-glucose units linked mainly by glycosidic bonds (1 → 4) and are usually classified

according to their dextrose equivalency (DE). The DE of a maltodextrin determines its reducing capacity and is inversely related to its average molecular weight. MD was mainly chosen as it provided excellent oxidative stability to encapsulated oil (Madene *et al*., 2006; Gharsalloui *et al*., 2007). Then the blend of biopolymers (MG-MD) utilized in this work as wall material retained and protected the oil into the microcapsules very well, and results were exhibited in higher EE%. The wall material to core ratio is another parameter to consider. It is generally accepted that a wall material to core ratio between 1:1 and 4:1 (w/w) should be suitable for most applications in dried food by spray-drying (Roccia *et al*., 2014).

### *4.4 Linoleic acid content in oil microencapsulated (LAC, %)*

Linoleic acid content in sesame oil was determined by gas chromatography before microencapsulated, which was found in 50.4%. After of the process of microencapsulation by spray drying, LAC% on the sesame oil varied from 47.68 to 50.39% (Table 2), these values were very similar at linoleic acid content original of the oil, therefore the independent variables not significantly influenced, although the combination between wall material to core ratio Wa:Co ($X_2$) with the drying air inlet temperature ($X_3$) was statistical significantly (Table 4).

Table 4. Regression analysis of variance and fitness of the models

| Source | Sum of squares | DF | Mean square | F value | p-Value |
|---|---|---|---|---|---|
| **LAC%** | | | | | |
| Model | 9.87 | 8 | 1.23 | 83.95 | <0.0001 |
| $X_1$ | 1.41 | 1 | 1.41 | 95.99 | <0.0001 |
| $X_2$ | 0.00093 | 1 | 0.00093 | 0.06 | 0.8027 |
| $X_3$ | 0.33 | 1 | 0.33 | 22.68 | <0.0001 |
| $X_1X_2$ | 1.95 | 1 | 1.98 | 132.79 | <0.0001 |
| $X_1X_3$ | 0.33 | 1 | 0.15 | 22.67 | <0.0001 |
| $X_2X_3$ | 3.3 | 1 | 3.3 | 224.27 | <0.0001 |
| $X_1^2$ | — | — | — | — | — |
| $X_2^2$ | 1.98 | 1 | 1.96 | 135.02 | <0.0001 |
| $X_3^2$ | 0.15 | 1 | 0.16 | 10.42 | 0.0037 |
| Residual error | 0.34 | 23 | 0.01 | | |
| Lack of fit | 0.33 | 18 | 0.02 | 11.95 | 0.0061 |
| C.V. % | 0.24 | — | — | — | — |
| Adeq Precision | 36.47 | — | — | — | — |
| **MC%** | | | | | |
| Model | 26.73 | 7 | 3.82 | 382.68 | <0.0001 |
| $X_1$ | 2.43 | 1 | 2.43 | 243.93 | <0.0001 |
| $X_2$ | 0.71 | 1 | 0.71 | 71.36 | <0.0001 |
| $X_3$ | 2.59 | 1 | 2.59 | 260.03 | <0.0001 |
| $X_1X_2$ | 4.69 | 1 | 4.69 | 470.15 | <0.0001 |
| $X_1X_3$ | — | — | — | — | — |
| $X_2X_3$ | — | — | — | — | — |
| $X_1^2$ | 8.39 | 1 | 8.39 | 840.84 | <0.0001 |
| $X_2^2$ | 0.44 | 1 | 0.44 | 43.78 | <0.0001 |
| $X_3^2$ | 2.43 | 1 | 2.43 | 243.23 | <0.0001 |
| Residual error | 0.24 | 24 | 0.01 | | |
| Lack of fit | 0.24 | 19 | 0.01 | 101.16 | <0.0001 |
| C.V. % | 2.68 | — | — | — | — |
| Adeq Precision | 61.79 | — | — | — | — |

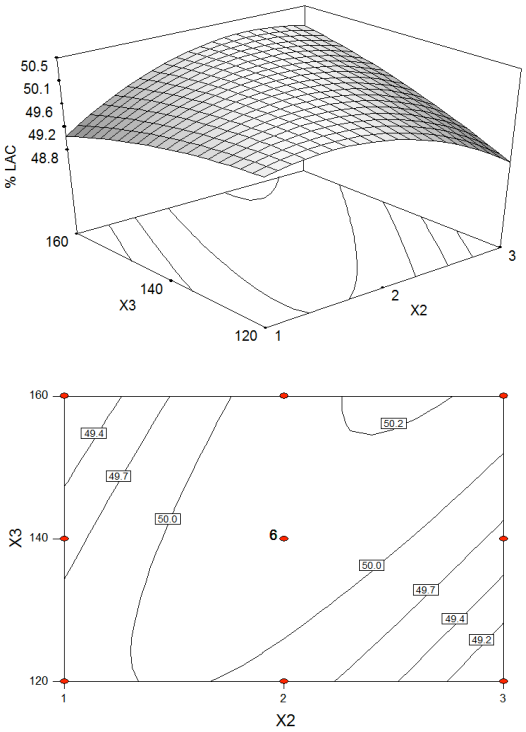


Fig. 4. The (a) 3D response and (b) 2D contour plots of the LAC % affected by volumetric dispersed phase ($X_1$) and wall material to core material ratio ($X_2$).

As seen in Fig. 4, the LAC% remained almost constant regardless of the values of $X_2$ and $X_3$. This is attributed to the blend of biopolymers as wall material, where the mesquite gum (MG) and maltodextrin DE-10 (MD) (MG66%-MD34% w/w) have high molecular weights that led to form a polymer matrix was able to retain the linoleic acid in the oil microencapsulated, inferring that this matrix delay mechanisms diffusive oxygen into the microcapsules. In addition, the glass transition of the wall material plays an important role in the linoleic acid content. Apparently the glass transition temperature of the wall material suggest that this is in a glass state due at lower moisture content in the microcapsules (Table 2) so the oxygen diffuses into the matrix is slow showing a larger resistance to oxygen diffusion. However, we cannot further discuss the effect of the diffusion rate of oxygen on the linoleic acid content because there is no information about the diffusion coefficient of oxygen through the dehydrated layer of matrix, or the rate constant of the oxidation kinetics of linoleic acid in microcapsules.

Some works has found that drying air inlet temperature is a parameter important that determine the quality and functionality of the microcapsules produced by spray-drying. Gharsallaoui *et al.* (2007) described in his review that is important to determine an appropriate drying temperature to avoid damage of the dried food product. There is a directly proportional drying temperature to the evaporation rate and inversely with the final water content of the powder. At low drying temperature, the formation of microcapsules with high density membranes, high water content, poor fluidity and easiness of agglomeration is caused by the low drying rate. In contrast, an excessive evaporation that occurred at high inlet temperatures results in cracks in the matrix inducting subsequent premature release, and therefore a degradation of encapsulated bioactive compounds. In this work, the drying air inlet temperature not influenced in the LAC% because of the range of temperature utilized not caused loss of linoleic acid microencapsulated; this may be explained because the obtained matrix could be found in a glassy state.

## *4.5 Moisture content (MC, %)*

The moisture content of the microcapsules varied from 2.63 to 5.64% (Table 2). Most of MC% values obtained through the experimental design were under the minimum moisture specification of dried powder in the food industry which is between 3 and 4 g/100 g water (Klaypradit and Huang, 2008; Huang *et al.*, 2014). The MC% variable was mainly affected by quadratic effect of the volumetric dispersed phase (X1) (Table 4). Although, this variable by itself not shows a clear trend with respect to moisture content (Fig. 5), the MC% values that are above 4% wt. could be due to the presence of the maltodextrin molecules in the wall material that be bind the water molecules makes it difficult water to diffuse. In addition, possibly the drying air inlet temperature not generated an increase in temperature gradient between the air and the sprayed product resulting in a lower heat and mass transference that decreased the water evaporation rate. This result also was related to the decreased rate of crust formation in the matrix, which hinders the water diffusion from sprayed particles. Therefore, the moisture content was not a critical parameter because the most of the microcapsules produced had low moisture content and were within the range that is considered safe for avoiding microorganism growth and the development of product alteration related to the initial moisture content.

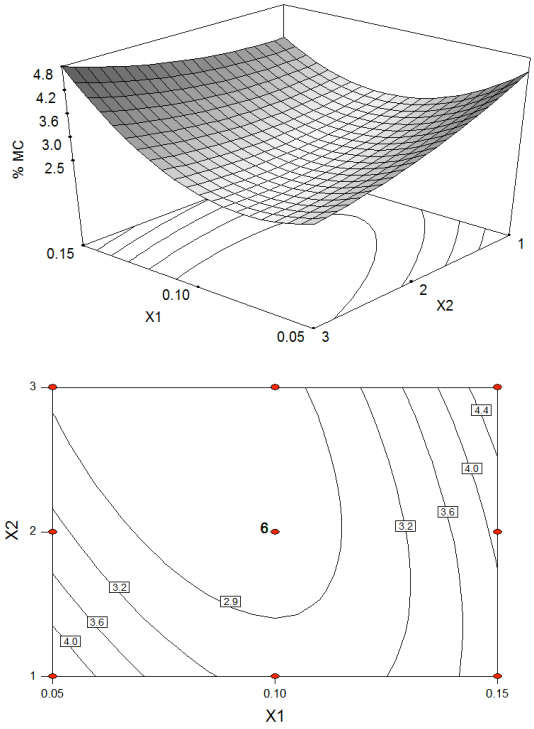


Fig. 5. The (a) 3D response and (b) 2D contour plots of the MC % affected by volumetric dispersed phase ($X_1$) and wall material to core material ratio ($X_2$).

### 4.6 *Determination of optimal condition*

The process parameters were optimized to achieve maximum efficiency encapsulation, maximum linoleic acid content microencapsulated, minimum surface oil on the microcapsules, and acceptable moisture content. The Derringer's desirability function was employed to obtain the optimum process variables. In order to maximize the microencapsulation of sesame oil, the following constraints have been taken into account (1) volumetric dispersed phase $\phi_{O/W}$) (0.05-0.15), (2) Wall material to core ratio (1:1-3:1) and (3) drying air inlet temperature (120-160 °C), were set for maximum desirability. The methodology of desired function was applied and the optimum level of various process variables were obtained to indicate that 0.05 of volumetric dispersed phase, 2.59:1 of wall material to core ratio and 154.04 °C drying air inlet temperature gives 11.80% surface oil, 88.02% encapsulation efficiency, 50.02% linoleic acid content in sesame oil microencapsulated and 2.99% (d.b.) moisture content with overall desirability value of 0.874.

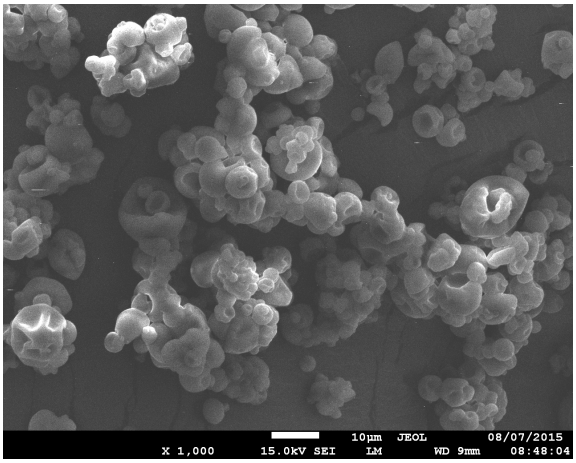


Fig. 6. Micrograph of the surface topology of microcapsules produced under the optimal conditions: volumetric dispersed phase of 0.05 ($X_1$), wall material to core material ratio of 2.59:1 ($X_2$) and drying air inlet temperature 154.04 °C ($X_3$).

### 4.7 *Morphology of the microcapsules*

Fig. 6 shows the optimally produced microcapsules, these particles exhibited various sizes (less than 20 $\mu$m). The microcapsules had irregular shape, concave and shriveled surface, which are the typical characteristics of microcapsules produced by spray drying. Most of the microcapsules showed no apparent cracks or fissures, ensuring a low permeability for oils and volatile compounds offering better protection and core retention, this type of morphology also has been observed by Tonon *et al*. (2011) and Kha *et al*. (2014) who observed that the formation of hollow particles is an usual characteristic of spray drying process (Ré, 1998).

The heat transfer rate and the water diffusion rate from surface to the core of the droplets as well as the wall material affected the microstructure of spray-dried matrices. According to Nijdam and Langrish (2006), it can be explained by the formation of a "vacuole" inside the particles, immediately after the crust development. This crust inflates when the particle temperature exceeds the local ambient boiling point and the vapour pressure within the vacuole rises above the local ambient pressure.

## Conclusions

This study was found that the statistical analysis was a valuable method for evaluating the effects of the volumetric dispersed phase, the Wa:Co ratio and the

drying air inlet temperature of the microencapsulation of sesame oil process. A volumetric dispersed phase of 0.05, Wa:Co ratio of 2.59:1, and drying air inlet temperature of 154.04ºC were suggested to be the optimum conditions for the microencapsulation by spray drying of sesame oil, providing the highest efficiency encapsulation (88.02%) and retention of linoleic acid (50.02%) in microcapsules values in the selected ranges. Therefore, it can be concluded that the high efficiency encapsulation of the sesame oil microcapsules and high retention of acid linoleic could be used as parameters to obtain functional food in the food industry.

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