Microencapsulation of flaxseed oil by spray drying: Influence of process conditions and emulsion properties

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ABSTRACT

The objective of this work was to evaluate the influence of inlet air temperature and emulsion composition (total solid content and oil concentration), as well as its associated properties (viscosity and droplet size), on the microencapsulation of flaxseed oil by spray drying, using gum Arabic as wall material. Coarse emulsions were prepared by blending the flaxseed oil and the gum Arabic solution, using a rotor-stator blender. They were then characterized with respect to droplet size and viscosity. Microencapsulation was performed in a laboratory scale spray dryer. Seventeen tests were made, according to a 2^3 central composite design. Independent variables were: inlet air temperature (138 – 202ºC), total solid content (10 – 30%) and oil concentration with respect to total solids (10 – 30%). Encapsulation efficiency and lipid oxidation were analysed as responses. Higher oil concentration and lower solid content led to the formation of larger oil droplets and less viscous emulsions. Encapsulation efficiency was negatively affected by oil concentration and positively influenced by total solid content. Lipid oxidation was higher for the emulsions containing higher oil concentration and lower solid content, which was attributed to the lower encapsulation efficiency obtained at these conditions. Peroxide values also increased when inlet air temperature increased.

Keywords: Microencapsulation; spray drying; emulsion properties; encapsulation efficiency; lipid oxidation.

INTRODUCTION

Microencapsulation of oils in a polymeric matrix is an alternative that has been used by several researchers in order to protect unsaturated fatty acids against lipid oxidation, thus increasing their shelf life. Besides protecting oils against oxidative damage, it also offers the possibility of controlled release of lipophilic functional food ingredients and can be useful for supplementation of foods with polyunsaturated fatty acids.

In the case of foods, the most common procedure for microencapsulation is spray drying. It involves the atomization of emulsions into a drying medium with high temperature, resulting in a very fast water evaporation, which results in a quick crust formation and in a quasi-instantaneous entrapment of the core material [1].

Gum Arabic is one of the most traditional wall materials used in the microencapsulation of oils and flavors. It is a natural gum with good emulsifying properties, since it has a little protein content in its composition. Moreover, it exhibits high solubility and low viscosity in aqueous solution when compared to other hydrocolloid gums [2], which facilitates the spray drying process.

In the last years, special attention has been given to the studies aiming at improving the encapsulation efficiency during spray drying of food flavors and oils, by minimizing the amount of unencapsulated oil present at the surface of powder particles and thus preventing lipid oxidation and volatile losses, and extending product’s shelf life. According to Jafari et al. [3], the main factors that affect encapsulation efficiency of microencapsulated oils and flavors are: the type of wall material, the properties of the core materials (concentration, volatiliy), the characteristics of the infeed emulsion (total solids, viscosity, droplets size) and the conditions of the spray drying process (atomization type, inlet air temperature, air flow, humidity). Thus, it is important to optimize the drying process, in order to obtain the minimal surface oil in the powder particles.

Microencapsulation of oils by spray drying has been used to protect unsaturated fatty acids against oxidation. Within the context of the functional foods, products supplemented with unsaturated fatty acids, such as

Omega-3, have been considered very beneficial to human health, mainly due to the cardiovascular diseases prevention promoted by these compounds.

Most of the works found in the literature about microencapsulation of unsaturated fatty acids-rich oils use fish oil as core material [4, 5, 6]. On microencapsulation of flaxseed oil by spray drying, however, very few information is available.

Thus, the objective of this work was to study the influence of inlet air temperature and emulsion composition (total solid content and oil concentration), as well as its associated properties (viscosity and droplet size), on the microencapsulation of flaxseed oil by spray drying, using gum Arabic as wall material. Encapsulation efficiency and lipid oxidation were analyzed as responses.

MATERIALS & METHODS

Material

Flaxseed oil was purchased from Sabor da Terra (Bragança Paulista, Brazil), with the following fatty acid composition: 5.91% C16:0, 4.31% C18:0, 22.55 C18:1, 12.83% C18:2 and 53.55% C18:3.

Gum Arabic Instantgum BA$^{®}$ was kindly donated by Colloïdes Naturels Brazil (São Paulo, Brazil).

Methods

Preparation of emulsions

The wall material was dissolved in distilled water under magnetic agitation, one day before emulsification. Coarse emulsions were prepared by blending the flaxseed oil and the wall solution, using a rotor-stator blender (Ultra-Turrax IKA T18 Basic, Wilmington, USA), at 15500 rpm for 5 min.

Emulsion viscosity

Emulsion viscosity was measured through the determination of steady-shear flow curves, using a controlled stress Physica MCR301 rheometer (Anton Paar, Graz, Austria) with stainless steel plate-plate geometry with a diameter of 75 mm and a gap of 0.2 mm. Three flow ramps (up, down and up-cycles) were obtained in a range of shear stress corresponding to shear rates from 0 to 300s^{-1}, in order to eliminate any possible thixotropy effect. Trials were performed in triplicate, using a new sample for each repetition. Rheograms were analyzed according to empirical models and viscosity was calculated as the relationship between shear stress and shear rate.

Emulsions droplet size

The droplet size distributions of the feed emulsions were determined by optical microscopy, using a Jenaval optical microscope (Carl Zeiss, Oberkochen, Germany). The Sauter mean diameter (D_{32}) was calculated from 500 droplets using the image processing software ImageJ 1.38x, according to Equation (1):

$$D_{32} = \frac{\sum_i z_i D_i^3}{\sum_i z_i D_i^2} \tag{1}$$

Where z_i is the number of droplets with diameter D_i.

Microencapsulation by spray drying

Spray drying process was performed in a laboratory scale spray dryer LabPlant SD-05 (Huddersfield, England), with a 1.5 mm diameter nozzle and main spray chamber of 500 mm × 215 mm. The emulsion was fed into the main chamber through a peristaltic pump, feed flow rate was 12 g/min, drying air flow rate was 73 m^3/h and compressor air pressure was 0.06 MPa.

A rotatable central composite design was used to perform the tests for the microencapsulation of flaxseed oil, considering three factors (independent variables): inlet air temperature (138 – 202°C), total solid content (10 – 30% w/w) and oil concentration with respect to total solids (10 – 30% w/w). Five levels of each variable were chosen for the trials, including the central point and two axial points, giving a total of 17 combinations. The following polynomial equation was fitted to data:

$$y = \beta_0 + \beta_1 x_1 + \beta_2 x_2 + \beta_3 x_3 + \beta_{11} x_1^2 + \beta_{22} x_2^2 + \beta_{33} x_3^2 + \beta_{12} x_1 x_2 + \beta_{13} x_1 x_3 + \beta_{23} x_2 x_3 \tag{2}$$

Where β_n are constant regression coefficients; y is the response (encapsulation efficiency or lipid oxidation), and x_1, x_2 and x_3 are the coded independent variables (inlet air temperature, solid content and oil concentration, respectively).

The analysis of variance (ANOVA), test for the lack of fit, determination of R^2 and the generation of three-dimensional graphs were carried out using the Statistica 7.0 software (StatSoft, Tulsa, USA).

Encapsulation efficiency

Surface oil was measured by extraction with hexane, according to the method used by Bae and Lee [7]. Total oil was assumed to be equal to the initial oil, since preliminary tests revealed that all the initial oil was retained, which was expected, considering that flaxseed oil is not volatile. Encapsulation efficiency (EE) was calculated as follows:

$$EE = \left(\frac{TO - SO}{TO} \right) \times 100 \tag{3}$$

Where *TO* is the total oil content and *SO* is the surface oil content.

Lipid oxidation

Lipid oxidation was evaluated by determination of the peroxide value. The oil was extracted according to the method described by Partanen et al. [8]. Peroxide value was determined spectrophotometrically, according to the IDF standard method [9] with some modifications. A portion of the extraction medium (600 μl) was added to 2.8 ml of a chloroform/methanol (7:3) mixture. For color formation, 30 μL of an ammonium thiocyanate/iron (II) chloride solution (1:1) were added. The sample was vortexed, reacted in the dark for 20 minutes, and absorbance was measured at 500 nm. Hydroperoxide concentrations were determined using a Fe^{+3} standard curve with iron concentration varying from 1 to 25 μg [9].

RESULTS & DISCUSSION

Emulsions characterization

The viscosity and the droplets mean diameter of the emulsions prepared with different solid content and oil concentration are presented in Table 1.

The most appropriate mathematical model for describing the flow characteristics was the Newton model, according to which viscosity is constant with shear rate. Emulsion viscosity increased as total solid content increased, which was expected, since gum Arabic is generally used as a thickening agent in foodstuffs, making them more viscous. According to Williams and Phillips [2], gum Arabic solutions are Newtonian fluids, whose viscosity exponentially increases with solution concentration.

The increase in oil concentration resulted in lower emulsion viscosities. This can be attributed to the lower amount of gum Arabic present in the emulsions formed with higher oil concentration, for the same total solid content, resulting in a slightly less pronounced thickening effect and thus in less viscous emulsions.

Table 1. Viscosity and droplets mean diameter of emulsions prepared with different solid content and oil concentration.

Solid content (%)	Oil concentration (%)	Viscosity (×10^3 Pa.s)	D_{32} (μm)
10	20	4.41 ± 0.06^a	4.41 ± 0.05^g
14	14	8.57 ± 0.05^c	3.20 ± 0.04^c
	26	6.99 ± 0.02^b	3.55 ± 0.04^d
20	10	23.33 ± 0.78	3.87 ± 0.04^e
	20	19.89 ± 1.09^e	4.16 ± 0.05^f
	30	15.89 ± 0.77^d	4.77 ± 0.06^h
26	14	50.35 ± 1.19^g	2.88 ± 0.04^b
	26	40.31 ± 0.20^f	3.28 ± 0.08^c
30	20	85.36 ± 1.51^h	2.27 ± 0.05^a

Droplets mean diameter varied from 2.27 to 4.77 μm. The increase in total solid content resulted in smaller droplets size, for the same oil concentration. These results can be related to the emulsions viscosity. As previously discussed, emulsions produced with higher solid content were more viscous. This higher viscosity reduces the rate at which particles sediment or cream, resulting in better emulsion stabilization and thus avoiding droplets coalescence [10].

On the other hand, higher oil concentrations led to higher mean diameters. The increase in oil concentration implies in lower gum Arabic content, for the same total solid content. Since gum Arabic has emulsifying properties, the lower concentration of this wall material may have resulted in a less efficient emulsification.

An emulsifier is a surface-active substance that is capable of adsorbing to an oil-water interface and protecting the droplets from flocculation and/or coalescence [10]. Thus, the decrease of gum Arabic content may have promoted higher droplets coalescence, resulting in the formation of larger droplets.

Response surface analysis

Encapsulation efficiency and lipid oxidation values obtained for each trial are shown in Table 2.

Table 3 shows the regression coefficients for the coded second-order polynomial equation, the F values and the determination coefficients (R^2). Some non-significant terms were eliminated and the resulting equations were tested for adequacy and fitness by the analysis of variance (ANOVA). The fitted models were suitable, showing significant regression, low residual values, no lack of fit and satisfactory determination coefficients.

Table 2. Encapsulation efficiency and lipid oxidation for the 17 trials of the experimental design.

Trials	Inlet air temperature (ºC)	Solid content (%)	Oil concentration (%)	Encapsulation efficiency (%)	Lipid oxidation (meq peroxide/kg oil)
1	150 (-1)	14 (-1)	14 (-1)	72.7 ± 1.0	0.047 ± 0.004
2	190 (+1)	14 (-1)	14 (-1)	73.1 ± 1.1	0.064 ± 0.006
3	150 (-1)	26 (+1)	14 (-1)	88.7 ± 0.3	0.027 ± 0.001
4	190 (+1)	26 (+1)	14 (-1)	88.9 ± 0.3	0.042 ± 0.002
5	150 (-1)	14 (-1)	26 (+1)	51.7 ± 0.3	0.066 ± 0.001
6	190 (+1)	14 (-1)	26 (+1)	53.2 ± 1.1	0.075 ± 0.006
7	150 (-1)	26 (+1)	26 (+1)	62.4 ± 2.0	0.037 ± 0.003
8	190 (+1)	26 (+1)	26 (+1)	60.5 ± 0.5	0.062 ± 0.001
9	138 (-1.68)	20 (0)	20 (0)	81.2 ± 0.2	0.038 ± 0.002
10	202 (+1.68)	20 (0)	20 (0)	77.5 ± 0.1	0.086 ± 0.002
11	170 (0)	10 (-1.68)	20 (0)	51.5 ± 1.4	0.080 ± 0.002
12	170 (0)	30 (+1.68)	20 (0)	84.6 ± 0.2	0.047 ± 0.001
13	170 (0)	20 (0)	10 (-1.68)	92.0 ± 0.1	0.017 ± 0.001
14	170 (0)	20 (0)	30 (+1.68)	58.6 ± 1.5	0.106 ± 0.009
15	170 (0)	20 (0)	20 (0)	81.1 ± 0.1	0.022 ± 0.002
16	170 (0)	20 (0)	20 (0)	75.1 ± 0.7	0.031 ± 0.002
17	170 (0)	20 (0)	20 (0)	77.7 ± 0.1	0.028 ± 0.002

Table 3. Coded second-order regression coefficients for encapsulation efficiency and lipid oxidation.

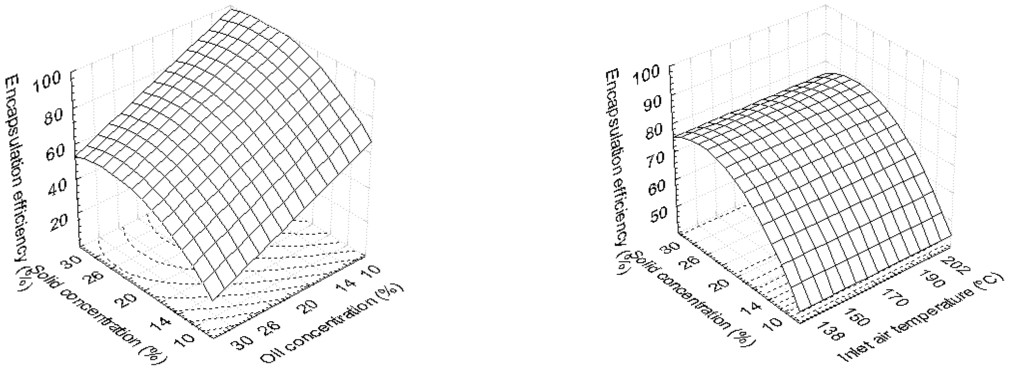
Coefficient	Encapsulation efficiency (%)	Lipid oxidation (meq peroxide/kg oil)	Coefficient	Encapsulation efficiency (%)	Lipid oxidation (meq peroxide/kg oil)
β_0	75.23	0.028	β_{33}	N.S.	0.010
β_1	N.S.	0.011	β_{12}	N.S.	N.S.
β_2	7.72	–0.010	β_{13}	N.S.	N.S.
β_3	–11.13	0.015	β_{23}	N.S.	N.S.
β_{11}	N.S.	0.010	R^2	0.927	0.808
β_{22}	–3.82	0.010	F	55.0	7.3

N.S. Non-significant.

Encapsulation efficiency

According to Tables 2 and 3, encapsulation efficiency varied from 51 to 92% and was significantly influenced by total solid content and oil concentration. Figure 1 shows the influence of the independent variables on this response.

Oil concentration was the factor that most affected encapsulation efficiency, showing a negative effect on this response. The same behavior was observed by Huynh et al. [11] in the microencapsulation of lemon myrtle oil, using modified starch + maltodextrin and whey protein concentrate + maltodextrin as wall materials. Tan et al. [5] also verified that high oil loadings resulted in lower process yield and lower encapsulation efficiency for microencapsulated fish oil by spray drying. The poorer retention related to higher oil loads can be attributed to the greater amount of core material close to the drying surface, which makes short the diffusion path length to the air/particle interface, thus increasing the surface oil content.



(a) (b)

Figure 1. Response surface for encapsulation efficiency of particles produced: (a) at inlet air temperature of 170ºC; (b) with total solid content of 20%.

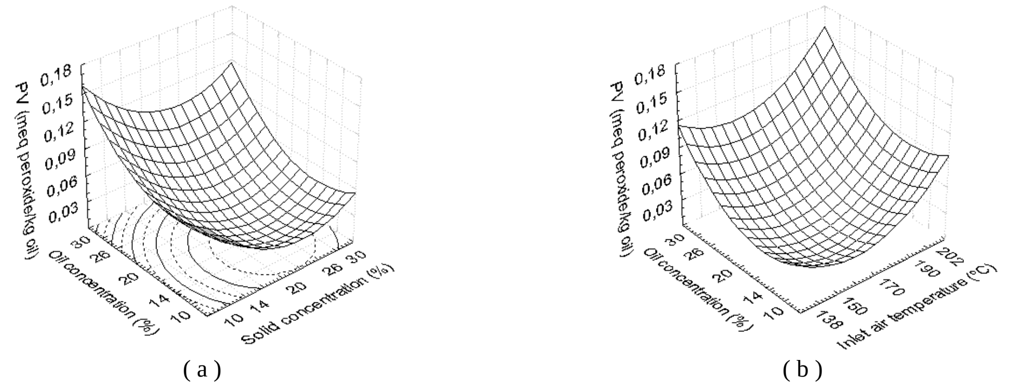
The effect of oil concentration on the encapsulation efficiency may also be related to the emulsion droplet size, which was lower for higher oil content. According to Jafari et al. [3], the higher surface oil in the particles produced from emulsions with larger droplets can be attributed to the droplets breakdown during atomization. Moreover, lower oil content resulted in higher emulsion viscosity, which reduces the oil droplets diffusion inside atomized droplet and makes difficult the oil migration to the particle surface.

Total solid content had a positive effect on the encapsulation efficiency, i.e., the increase in solid content resulted in higher encapsulation efficiency. Higher solid content implies in shorter time to form a crust, making difficult the oil diffusion to the drying particle surface. Moreover, increasing total solids leads to the increase of emulsion viscosity, reducing the circulation movements inside the droplets and, thus, resulting in a rapid skin formation [3].

The higher encapsulation efficiency shown by particles produced from emulsions with higher solid content may also be attributed to the emulsion droplets size, since increasing solid content led to the production of droplets with lower diameters. As previously discussed, the smaller the emulsion droplet, the higher is the encapsulation efficiency, which explains the results obtained.

Lipid oxidation

The influence of inlet air temperature, solid content and oil concentration on the lipid oxidation is shown in Figure 2.



(a) (b)

Figure 2. Response surface for lipid oxidation of particles produced: (a) at inlet air temperature of 170ºC; (b) with total solid content of 20%.

Lipid oxidation was affected by all the independent variables. In general, lower solid content and higher oil concentration led to higher peroxide values. This can be related to the lower encapsulation efficiency obtained at these conditions, which leads to poorer oil protection against oxidation. The lower the encapsulation efficiency, the higher is the amount of oil present in the particles surface. This non encapsulated oil, when in contact with the oxygen, is much more susceptible to lipid oxidation than the encapsulated one.

The test that resulted in the highest encapsulation efficiency (91.97%) was the same that provided the lowest lipid oxidation (0.017 meq peroxide/kg oil) (Table 2). On the other hand, the highest peroxide value (0.106 meq peroxide/kg oil) was obtained for the powder produced with 20% solids and 30% oil, condition in which the feed emulsion had the highest droplet mean diameter. This indicates that the emulsion droplet size also plays an important role on the protection against lipid oxidation in oils microencapsulated by spray drying.

Peroxide value also increased with increasing drying temperatures. The use of higher inlet air temperatures provides more energy available for the lipid oxidation process, favoring peroxides formation. Thomsen et al. [12] observed strong temperature dependence for quality deterioration of milk powders, verifying an Arrhenius type dependence of lipid oxidation on temperature. Serfert et al. [13] observed that hydroperoxide content of microencapsulated fish oil was three times higher for the powder produced at inlet/outlet temperatures of 210/90ºC, when compared to that produced at 160/70ºC.

CONCLUSION

The best conditions for flaxseed oil encapsulation, aiming at achieving high encapsulation efficiency and low lipid oxidation, were: inlet air temperature of 140-170ºC, total solid content of 26-30% and oil concentration of 10-14%. The presence of oil in the particles surface is very detrimental to powder quality, since it can lead to lipid oxidation. This work showed that the encapsulation efficiency directly depends on the feed emulsion properties (viscosity and droplet size). Thus, the study of these properties is essential for obtaining a successful microencapsulation and thus, particles of good quality.

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