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Polymeric microencapsulation of pequi oil: preparation and characterization

Rachel Menezes CASTELO[1], Luana Carvalho da SILVA[1], Herllan Vieira de ALMEIDA[2], Huai Nan CHENG[3], Atanu BISWAS[4], Hilton César Rodrigues MAGALHÃES[5], Roselayne Ferro FURTADO[5]*

Abstract

Microencapsulation is often used to protect an unstable bioactive ingredient from the actions of external factors. In our work, we have found biobased polymers to be particularly suited as polymeric microencapsulants. Thus, pequi oil (from *Caryocar coriaceum*) was encapsulated by ionic gelation through polyelectrolyte complex formation between chitosan and alginate. The process of microparticle formation was studied, and the formation of the polymeric emulsion and microparticles was fully characterized. The influence of freeze-drying and oven-drying on the characteristics of the microparticles was also investigated. The hydrophilic–lipophilic balance (HLB) was utilized as a testing protocol to prepare a stable emulsion for microencapsulation, with the HLB of 10.2 showing the best stability. The encapsulation efficiency and loading capacity were 96.2 and 37.0%, respectively. Oven-dried particles showed a smaller particle size and a lower degree of sphericity and swelling than freeze-dried particles. Furthermore, freeze-dried microparticles had a lower percentage of oleic acid than those dried in an oven. This systematic approach (involving preparation, characterization, and optimization) should be applicable to the polymeric microencapsulation of other unstable bioactive ingredients for food, cosmetic, and pharmaceutical applications.

Keywords: alginate; *Caryocar coriaceum*; chitosan; freeze-drying; HLB; microencapsulation; oven-drying; pequi oil.

Practical Application: The oven-dried microparticles maintained better the chemical characteristics of pequi oil.

1 INTRODUCTION

Microencapsulation is a process of enclosing an active substance (core) within a protective coating (shell) to form small particles or capsules (Bakry et al., 2016; Paulo & Santos, 2017). It can provide various benefits for the core material, such as protection from environmental factors, controlled release, improved stability, enhanced solubility, reduced volatility, and masking of undesirable properties. The selection of the shell material is a crucial factor for the success of microencapsulation. In recent years, there has been an increasing interest in using biobased polymers as shell materials for microencapsulation. Biobased polymers are derived from renewable biological sources, such as plants, animals, or microorganisms (Cheng & Gross, 2020; Simões et al., 2017). They can offer several advantages over synthetic materials, such as environmental friendliness, biocompatibility, biodegradability, low toxicity, low cost, and availability.

The edible oil from pequi fruit (*Caryocar coriaceum* Wittm), commonly known as pequi oil, is often used in folk medicine for anti-inflammatory, wound healing, and gastroprotective remedies (Saraiva et al., 2011). This oil contains vitamins A and E, carotene, and fatty acids such as oleic (omega-9), palmitic, stearic, linoleic (omega-6), and linolenic (omega-3). Due to its composition, this oil is sensitive to adverse environmental conditions such as heat, light, and oxygen (Oliveira & Scariot, 2010), which accelerate its oxidation and degradation. One way to protect the functional and nutraceutical properties of an unstable substance is to use microencapsulation with biobased polymers. Indeed, some studies have been reported on encapsulation of pequi oil (Alexandre et al., 2019; Comunian et al., 2020; Justi et al., 2018; Oliveira et al., 2018) using polymeric encapsulants such as gelatin and gum Arabic (Justi et al., 2018), cashew gum and gelatin (Alexandre et al., 2019), whey protein isolate (Comunian et al., 2020), and whey protein isolate, maltodextrin, and inulin (Oliveira et al., 2018).

Ionic gelation is an efficient and known encapsulation technique performed under room temperature and pressure (Menin et al., 2018). Homogeneous and wet particles may be obtained using this technique in combination with vibration nozzle technology. During the encapsulation process, vibrational energy with a controlled frequency and amplitude is superimposed onto the oil emulsion, which breaks up into small droplets of approximately equal size when extruded by a nozzle to form spherical particles. As the drops come into contact with monovalent or divalent cations, particles are formed by the gelling process (Dorati et al., 2013). Calcium chloride is commonly used and was chosen to promote the ionic gelation of alginate. Chitosan may be used as a reinforcement polymer

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[1]*Universidade Estadual do Ceará, Centro de Ciências e Tecnologia, Fortaleza, Ceará, Brazil.*
[2]*Universidade Estadual do Ceará, Rede Nordeste de Biotecnologia, Fortaleza, Ceará, Brazil.*
[3]*Southern Regional Research Center, New Orleans, Louisiana, United States of America.*
[4]*National Center for Agricultural Utilization Research, USDA Agricultural Research Service, Peoria, Illinois, United States of America.*
[5]*Embrapa Agroindústria Tropical, Fortaleza, Ceará, Brazil.*
*Corresponding author: roselayne.furtado@embrapa.br

for the alginate-based matrix. The polyelectrolytic complex between the protonated amino groups of chitosan and alginate carboxylate groups can give the particles more stability to variations in pH and greater efficiency in the controlled release of the active ingredients (Yadav et al., 2018). The alginate and chitosan polyelectrolyte complex favors the encapsulated agent's release at pH conditions similar to intestinal pH (Abbaszadeh et al., 2014) and facilitates its application as a functional additive for food or drug.

Using appropriate analytical methodologies and testing protocol to optimize the process for encapsulation is highly desirable. For example, the hydrophilic–lipophilic balance (HLB) (ICI Americas, 1984) can be an effective testing protocol to prepare a stable emulsion for microencapsulation. Moreover, no one has compared freeze-drying and oven-drying methods in combination with microencapsulation of pequi oil by ionic gelation. Drying methods can modify the morphological, functional, and release characteristics of the particles (Otálora et al., 2018; Raghavan, 2020). The drying process is very important for industrial applications because the dehydration of microparticles prevents the proliferation of microorganisms, enhances storage stability, and facilitates transport by reducing the volume.

In this work, the process for microencapsulation of pequi oil was optimized through the use of appropriate analyses and HLB testing protocol to study the emulsion formation and the influence of two drying methods (freeze-drying and oven-drying), particularly with respect to the morphology, physical, chemical, and physicochemical characteristics of the pequi oil microparticles. The approaches used here should be applicable to the microencapsulation of other unstable bioactive ingredients.

2 MATERIALS AND METHODS

2.1 Materials

Sodium alginate (medium viscosity) and calcium chloride were obtained from Dinâmica®, Brazil. Chitosan (low molecular weight, 75–85% deacetylation) and Span 80 surfactant were obtained from Sigma-Aldrich, St. Louis, MO, USA. Tween 80 surfactant was purchased from Vetec, RJ, Brazil, and Span 20 from Chemical Dynamics, SP, Brazil.

2.2 Extraction and characterization of pequi oil

The pequi fruits were washed, sanitized (chlorine 200 ppm for 30 min), peeled, and subjected to a hammer pulper (capacity of 300 kg/h). For oil extraction, the pulps were heated to 45°C and centrifuged at 4,500 rpm for 15 min to separate the oil (Lima et al., 2019). The oil yield was calculated as a percentage of the mass of the pequi pulp.

The fatty acids were converted into fatty acid methyl esters, and the analysis was performed in a gas chromatography instrument with a flame ionization detector (Shimadzu, model GC2010 Plus, Kyoto, Japan), equipped with an automator (Shimadzu Model AOC-5000) as described by Silva et al. (2022).

2.3 HLB Testing Protocol for Emulsion Preparation

A solution of alginate (1.2% w/v) in distilled water was prepared under stirring for 24 h. Tween 80 or Tween 20 (according to Table 1) was added to the alginate solution and homogenized in an Ultra-Turrax® equipment (Digital T-25, IKA®). Pequi oil (2% w/v) was stored at room temperature (25°C), and Span 80 or Span 20 (according to Table 1) was added to the previous alginate solution and homogenized at 12,000 rpm in an Ultra-Turrax® rotor–stator for 5 min. The determination of Tween and Span in the emulsion was calculated by Equation 1 (ICI Americas, 1984):

$$\%A = \frac{100\left(HLB(req) - HLB(B)\right)}{HLB(A) - HLB\ (B)} \quad (1)$$

$$\%B = 100 - \%A$$

where:

A: the lipophilic surfactant (Span 80 or 20);

B: the hydrophilic surfactant (Tween 80 or 20);

HLB (req): the HLB value required for the oil.

The mass of Tween 80, Tween 20, Span 80, or Span 20 was calculated according to the HLB values: *HLB = 10.2, HLB = 10.2, HLB = 9.1, HLB = 9.0, HLB = 11.2* [15] (Table 1).

2.4 Characterization of emulsions

2.4.1 Turbidity analysis

The turbidity values (TV) were obtained by UV–visible spectrophotometry at 600 nm wavelength. The TV were calculated according to Equation 2 (Pearce & Kinsella, 1978):

$$TV = \frac{2.303 \times A\lambda \times fd}{l} \quad (2)$$

Table 1. Composition of O/W type of emulsions prepared with pequi oil (*Caryocar coriaceum*), alginate (1.2%), and surfactant.

Emulsions	Tween 80/g	Span 80/g	Tween 20/g	Span 20/g	Total surfactant concentration (%)
HLB 10.2 (1)	0.55	0.45	-	-	1
HLB 10.2 (2)	-	-	0.20	0.80	1
HLB 9.1 (3)	0.45	0.55	-	-	1
HLB 9.0 (4)	-	-	0.95	0.05	1
HLB 11.2 (5)	0.65	0.35	-	-	1
Control (6)	-	-	-	-	-

Where:

A_λ: the emulsion absorbance value at 600 nm;

fd: the dilution factor of the emulsion;

l: the optical path of the cuvette.

2.4.2 Viscosity analysis

Analysis of the viscosities of the emulsions (alginate–pequi oil) was performed using a HAAKE MARS III rheometer (Thermo Scientific, Pittsburgh, PA, USA). The viscosity was determined at a shear rate of 100 s^{-1}.

2.4.3 Analysis of zeta potential and particle size

For the analysis of zeta potentials and average particle sizes of the emulsions, 1 mL of the dispersions, previously stirred, was diluted in 100 mL of deionized water, homogenized, and analyzed in a Zetasizer Nano ZS equipment (Malvern, USA). Each determination was performed in triplicate.

2.5 Production and characterization of microspheres

A 100-mL solution of 1.2% (w/v) sodium alginate was prepared, and 2 g of pequi oil was added, followed by mechanical stirring for 30 min. The emulsion was dripped into a solution of 1.3% (w/v) calcium chloride and 1.2% (w/v) chitosan under stirring for 24 h (González-Rodríguez et al., 2002). The vibration nozzle encapsulation was performed with a Büchi B-395 encapsulator equipment (Essen, Germany) using a 120-µm drip nozzle (Castelo et al., 2020). The operating parameters included 300 V (electrostatic charge) and 200 mbar air pump pressure. The air pressure regulation system settings included 0.5 kPA, a flow rate of 1.7 mL min^{-1}, and a frequency of 1,800 Hz. The distance between the nozzle and the gelling bath was 20 cm (Castelo et al., 2020).

2.6 Encapsulation efficiency

Encapsulation efficiency (EE) was determined from total and surface oil content of wet particles according to Equation 3. Total oil refers to the total amount of oil (2% w/v) in 100 mL of emulsion. The surface oil content was determined by washing the particles with 10 mL of hexane and quantifying the oil present using a spectrophotometer at 450 nm. Previously, a calibration curve was established for pequi oil in hexane.

$$EE(\%) = \frac{(total\ oil - surface\ oil)}{total\ oil} \text{ x } 100 \quad (3)$$

2.7 Loading capacity

Loading capacity (LC) of the wet microparticles was determined by measuring the amount of oil loaded per unit mass of the carrier (Equation 4):

$$LC(\%) = \frac{total\ oil}{total\ mass}\ x\ 100 \quad (4)$$

where:

the total oil: the initial oil mass used;

the total mass: the mass of the particle (core + matrix).

2.8 Drying process

The spheres were subjected to dehydration using oven-drying (Tecnal, Brazil, Model TE-393/1) at 50 °C for 2.5 h and freeze-drying (M. Christ, Germany, Model 1-8 LSC basic) for 48 h. Before freeze-drying, the samples were frozen in an ultra-freezer (Sanyo, Japan, VIP Series-86 °C Ultra-Low, Temperature Freezer Model MDF-U33V).

2.9 Particle size analysis

The particle size was determined from images obtained using an optical microscope (Zeiss, Germany, Model Axio Imager A2). The micrographs were captured for 20 randomized microspheres. The measurements of transverse and longitudinal diameters were taken, and the medium size (MS) was calculated using the Ferret diameter (Zanetti et al., 2002) according to Equation 5:

$$MS = \frac{d + D}{2} \quad (5)$$

Where:

d: the smallest diameter of an inscribed circle;

D: the largest diameter of the circumscribed circle, both at the largest cross section of the particle.

2.10 Sphericity analysis

The degree of sphericity of the obtained particles was determined using the Riley method (Riley, 1941) according to Equation 6:

$$\Phi 0\ = \sqrt{\frac{d}{D}} \quad (6)$$

Where:

Φ_0: the sphericity;

d: the largest diameter of an inscribed circle;

D: the largest diameter of the circumscribed circle, both at the largest cross section of the particle.

2.11 Scanning Electron Microscopy

Micrographs were obtained using a scanning electron microscope (SEM; Quanta 450 FEG System: FEI Company, USA) with a scanning voltage of 15 kV. The samples were metalized

with a thin silver layer produced by a sputter coater (Emitech Model k550, Quorum Technologies, UK).

2.12 Swelling analysis

Twenty microspheres were separated and submerged in 15 mL of distilled water. The samples were maintained at room temperature (25 ± 2°C) for 24 h; the particles were separated, placed on filter paper to remove excess water from the surface, and then weighed. The swelling degree (SD) was determined according to Equation 7:

$$SD\% = \frac{Wt - Wo}{Wo}\ x\ 100 \quad (7)$$

Where:

Wt: the mass of the microspheres intumesced at time t;

Wo: the dry microsphere mass.

2.13 Statistical analysis

The data were analyzed using the Statistica 13 software (Dell, now TIBCO Software Inc., USA), and the values were compared using Student's t-test for dependent samples (when analyzed over time) and independent samples at the 5% level of significance.

3 RESULTS AND DISCUSSION

3.1 Characterization of pequi oil

The yield of pequi oil from pulp was 16.97% (w/w). This value agrees with the yield for aqueous extraction of pequi pulp oil reported by Mariano et al. (2009). The oil yield for the initial pulp mass, which in the artisanal method (heating for a long time) was less than 10% (w/w), reaches about 20% (w/w) using the technique of physical separation in an aqueous medium, corresponding to 76% of the oil in the pulp. Aqueous extraction of oil from plant materials at a mild temperature for food use has the advantage of not requiring organic solvents (Lima et al., 2019).

The fatty acid profile for pequi oil is shown in Table 2. The chromatographic analysis of the oil showed a predominance of unsaturated fatty acids (64.7%) when compared with saturated fatty acids (34.5%). Intake of unsaturated fatty acids (e.g., polyunsaturated fatty acids, PUFA), especially essential fatty acids, is highly recommended to prevent the risk of coronary heart diseases, such as hypertension and high cholesterol. However, unsaturation of the oil is a source of oxidative instability to processing and/or storage conditions at suboptimal lighting and temperatures. Oxidation reactions may occur quickly that lead to undesirable changes in sensory and nutritional properties (Da Matta, 2013).

Table 2. Chromatographic profile (obtained by gas chromatography) of fatty acids from pequi pulp oil (*Caryocar coriaceum*).

Fatty acid	% Composition (w/w) (%)
Oleic (C18:1)	61.40
Palmitic (C16:0)	32.81
Linoleic (C18:2)	2.32
Stearic (C18:0)	1.58
Palmitoleic (C16:1)	0.41
Linolenic (C18:3)	0.34
Arachidonic (C20:4)	0.20
Myristic (C14:0)	0.06
Heptadecanoic (C17:0)	0.05
Cis-10-heptadecenoic (C17:1)	0.05

3.2 Evaluation of pequi oil and alginate emulsions

Emulsions were prepared before the encapsulation process by considering the HLB of pequi oil between 10.5 and 11.75, as reported by Alvares (2016). The stability of emulsions with different HLB values (A, B, C, D, E, and F) was observed after 24 h. Intermediate values of HLB (Table 1) exhibited an opaque appearance and a homogeneous yellow color. In the dispersions B, E, and F, there were a clear separation of phases and the formation of a serum layer. The emulsion destabilization is related to several physical–chemical mechanisms: flocculation, coalescence, Ostwald ripening, and phase inversion. It is known that O/W emulsion is favored with HLB values in the range from 8 to 18 with the associated use of hydrophilic and lipophilic surfactants such as Tween and Span, respectively. Tween 80 has a longer aliphatic tail and is less hydrophilic than Tween 20. Similarly, Span 80 is also less hydrophilic than Span 20 (ICI Americas, 1984).

The emulsion with satisfactory stability for 24 h was ready for the subsequent encapsulation. In the absence of surfactants, the emulsion is destabilized, a fact that can harm the encapsulation process. If this happens, the flow to the equipment is altered, and nozzle clogging may occur during the process. Table 3 shows the analytical results of turbidity, zeta potentials, and viscosity values for alginate and pequi oil emulsions made with the surfactants.

There was no variation in the viscosity values of the emulsions. Viscosity plays an important role in liquid extrusion and on the adjustable parameters of the encapsulation equipment used in this work (i.e., frequency, amplitude, and injection nozzle size); it is considered a characteristic that mainly affects the size and morphology of the microparticles (Castelo et al., 2020). The zeta potential of the particles can be positive or negative in polarity, depending on the electrical potential being created by the presence of a charge on the droplet surface. Thus, the zeta potential value is used in the module as an important indicator of the probable physical stability of the formulation (Shah et al., 2014). According to the literature, a system is considered stable when it has an absolute value greater than 25 mV (Lieberman et al., 1989). When coagulation between particles occurs due to the Brownian motion, the stability is decreased, the repulsive force between particles is decreased, and the zeta potential value is also decreased. Except for the emulsion with HLB 11.2, all samples presented a zeta potential above 25 mV.

The TV is related to the stability of emulsions, with a high TV generally being attributed to a higher number of droplets or a reduction in their sizes. On the basis of the comparison of the turbidity test with other analyses (droplet size and zeta potential), HLB 10.2 was chosen as the criterion for the microencapsulation process.

3.3 Characterization of the dry particles

Alginate–pequi oil emulsion with HLB 10.2 (made with surfactants Tween and Span 80) was dripped into the chitosan and $CaCl_2$ solution. The polyelectrolyte complex formation between chitosan and alginate was reported in the literature as advantageous to the particle formation (Castelo et al., 2020; Yadav et al., 2018). A previous study showed that the particle formation without chitosan and drying at 50°C led to an oil overflow. The samples treated with chitosan and dried at 50°C were less spherical and displayed smoother characteristics.

The microspheres showed a clear morphological difference depending on the wall material and the drying method. Freeze-dried microspheres had a porous appearance and a degree of sphericity closer to 1 (for a perfect sphere, the value would be 1 (Zanetti et al., 2002). The porous characteristic is associated with the formation of ice crystals and their sublimation during the freezing process under reduced pressure.

The microspheres subjected to freeze-drying presented larger sizes relative to oven-dried samples (Figure 1). This is because heat drying causes the microspheres to shrink and the matrix wall to harden due to the evaporation of water by capillary action. This does not occur in freeze-dried spheres, as water changes from a solid to a vapor. Particle drying is desirable because it leads to improved storage properties due to the removal of water that hinders the proliferation of microorganisms and the reduction in the volume of the product (Raghavan, 2020). The reduction in particle size has always been an objective in the

Table 3. Zeta potentials, turbidity, viscosity, and particle size values of alginate and pequi oil emulsion with surfactants at different HLB values after 24 h.

Emulsion	Zeta potential (mV)	Turbidity	Viscosity (Pas)	Particle size (nm)		
				Behavior	Peak 1	Peak 2
HBL 10.2 (A)	-45.93 ± 0.47	179.63	0.160	Bimodal	900.15 ± 170.22	118.20 ± 42.15
HBL 10.2 (B)	-61.83 ± 1.85	74.63	0.170	Unimodal	1970.52 ± 88.65	-----
HBL 9.1 (C)	-46.60 ± 0.79	223.39	0.156	Bimodal	1831.58 ± 867.44	165.82 ± 59.27
HBL 9.0 (D)	-53.53 ± 1.04	232.37	0.165	Unimodal	2212.80 ± 823.61	-----
HBL 11.2 (E)	-14.63 ± 0.46	75.08	0.171	Bimodal	1791.41 ± 1130.59	127.66 ± 78.92
Control (F)*	-71.53 ± 0.15	200.82	0.157	Bimodal	1695 ± 253.46	432.00 ± 81.53

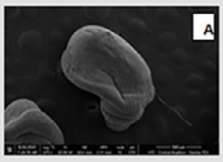
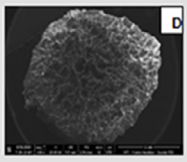
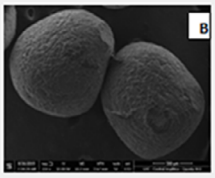
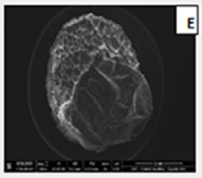
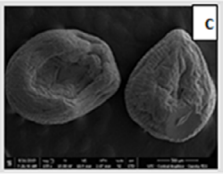
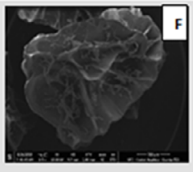
Oven-drying				Freeze-drying			
Sphere composition	Size (µm)	Sphericity	Microscopy	Sphere composition	Size (µm)	Sphericity	Microscopy
Alg/chit/oil	1059.18 ± 217.89^{a}	0.842 ± 0.12^{a}		Alg/chit/oil	1428.95 ± 95^{b}	0.940 ± 0.03^{a}	
Alg/oil	871.27 ± 27^{a}	0.918 ± 0.08^{a}		Alg/oil	1614.99 ± 10.96^{d}	0.973 ± 0.001^{a}	
Alg	744.77 ± 39.07^{c}	0.885 ± 0.04^{a}		Alg	1372.62 ± 127.90^{b}	0.866 ± 0.037^{a}	

Figure 1. Particle size, sphericity, and SEM micrographs of microparticles dried by two different drying methods, where "alg" is alginate, "chit" is chitosan, and "oil" is pequi oil. The microparticles of A, B, and C were submitted to oven-drying at 50 °C and of D, E, and F to freeze-drying. The same letters in the same column did not differ statistically ($p > 0.05$)

design of microencapsulated systems because smaller particles can pass through the gastrointestinal tract more efficiently and provide a larger surface area for the release of the bioactive compound (Agüero et al., 2017; Teixeira et al., 2014).

Deladino et al. (2008) evaluated the drying methods applied to microspheres containing natural antioxidants of the yerba mate *Ilex paraguariensis*, where oven-drying at 80 °C showed better results compared with freeze-drying because the freeze-dried microspheres showed a porous structure, which increased the external surface and escalated the exposure of the active compound. Teixeira et al. (2014) produced oven-dried alginate particles for applications in drug transport systems with more cohesive particles and smaller sizes.

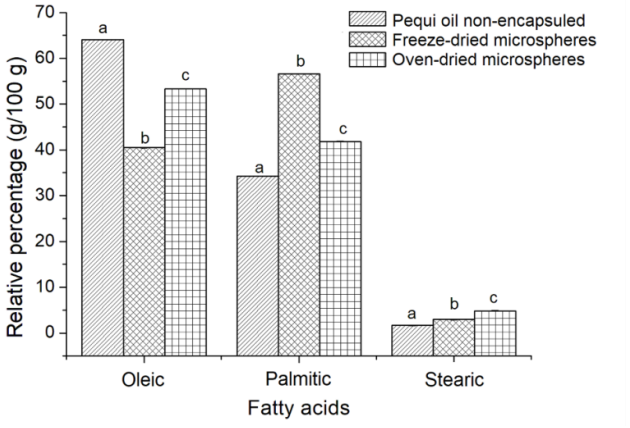
Regarding the fatty acid composition compared with the initial amount of pequi oil (Table 2), the drying methods influenced the saturation and unsaturation degree of the fatty acids present (Figure 2). The freeze-dried microparticles had a lower percentage of oleic acid than the oven-dried counterparts. This result agrees with previous studies that demonstrated that the porous structure and large surface area of the freeze-dried samples favor the easy penetration of oxygen and exposure to free radicals, thereby increasing lipid oxidation over time (Deladino et al., 2008). The oven-dried microparticles presented a more homogeneous and hardened surface structure, representing higher protection for the encapsulated material.

One of the most important characteristics of hydrophilic polymers with respect to their application in release systems is the swelling in water, which has a pronounced effect on the retention of the incorporated compound (Lopes et al., 2005). The freeze-dried spheres showed a higher swelling percentage than the dried spheres at 50°C (Figure 3). This fact is associated with the porous morphology of the freeze-dried spheres that facilitates the entry of water through the particle wall. Dorati et al. (2013) pointed out that after the drying of alginate microparticles by freeze-drying, these microparticles showed high percentages of rehydration in water and a simulated biological fluid (pH 7.4). Marfil et al. (2018) microencapsulated palm oil and evaluated the effect of freeze-drying on the spheres, obtaining good reproducibility and rehydration capacity of spheres. On the contrary, in this work, the microspheres dried by heating showed a lower SD due to the formation on their surface of a rigid and almost impermeable film. In this case, the solute present in the sample moves from the center to the surface of each particle, together with the water; on the surface, the water evaporates, and the solutes are deposited, closing the pores (Raghavan, 2020).

3.4 Encapsulation efficiency and loading capacity of wet microparticles

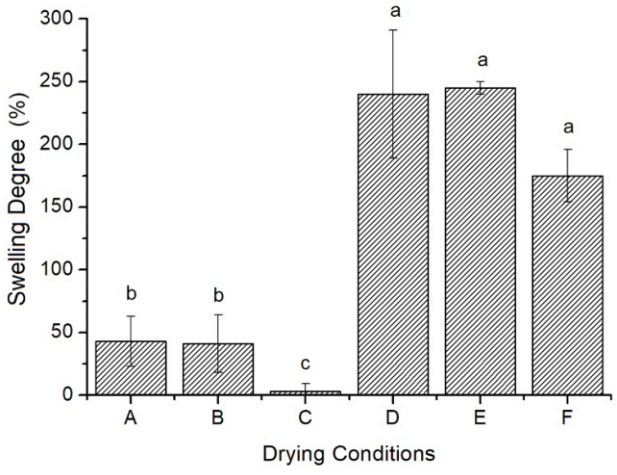
The EE and the LC are two important factors in evaluating the encapsulation process. The pequi oil EE of alginate and chitosan microspheres was 96.17%. On the contrary, the particles formed only with alginate showed an EE of 67.85%. Similar results have been reported in the literature performed with comparable levels of EE. Savic et al. (2022) reported an EE of 89.2% for antioxidants extracted by ultrasound-assisted with ethanol from orange peel and encapsulated in alginate–chitosan matrix. Another study by Gajic et al. (2021) with carotenoids extracted from orange peel using olive oil as a solvent showed an ample and successful oil loading in Ca-alginate beads. An EE of 89.5% was found in that work, and the separation of the solvent from oil enriched with carotenoids was not necessary. Sanches et al. (2020) produced nanoparticles with citronella essential oil using alginate–chitosan, and the EE was approximately 80%.

A desirable LC requires that the particle mass contains a relatively high amount of the compound of interest. Thus, particles with a low LC may not be viable because more encapsulated material would be needed. However, an LC above 50% is also not desirable because it reduces the protection of the matrix over the core (Shaddel et al., 2018). A high LC value also



[a]The same letters did not differ statistically in relation to non-encapsulated and encapsulated oil for each fatty acid ($p > 0.05$).

Figure 2. Relative percentage (g/100 g) of the major fatty acids (oleic, palmitic, and stearic) found in the pequi oil non-encapsuled and oven-dried and freeze-dried microspheres[a].



[a]The same letters did not differ significantly ($p > 0.05$).

Figure 3. Swelling percentage of the microspheres[a] by oven-drying at 50 °C and freeze-drying. Samples A, B, and C were oven-dried and are alginate + chitosan + oil, alginate + oil, and alginate, respectively. Samples D, E, and F were freeze-dried and are alginate + chitosan + oil, alginate + oil, and alginate, respectively.

suggests that the core material is close to the surface and can be quickly degraded or released onto the environment. In this work, microspheres with the alginate–chitosan matrix showed an LC of 37.04%, while microspheres formed only with alginate without chitosan presented an LC of 47.61%. A consequence of the higher LC value of the particles using only alginate as a matrix is the easy extravasation of the oil, as was verified during the drying of the particles at 50°C.

4 CONCLUSION

In this work, the HLB test protocol was used successfully to obtain more stable emulsions for encapsulation of pequi oil by ionic gelation; previously, it was more commonly used for encapsulation by spray-drying. The characterization methods permitted optimization to be performed. The drying methods influenced the characteristics of microparticles and the unsaturation degree of the fatty acids. The freeze-dried microparticles had a lower percentage of oleic acid than the oven-dried microparticles, probably due to a higher susceptibility of them to external factors. This is related to the final characteristics of the microparticles after each drying method, where the oven-dried particles were more hardened, while the freeze-dried particles showed a porous morphology, with greater sphericity and more rapid swelling. This analytical approach reported herein (involving the combination of various characterization methods, test protocols, and procedures) should be helpful for the future preparation and applications of microencapsulation of other unstable bioactive compounds.

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