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ORIGINAL ARTICLE

Synthesis and characterization of microalgae fatty acids or *Aloe vera* oil microcapsules

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Abstract

It's proposed a single methodology for the encapsulation of *Aloe vera* oil or microalgae fatty acids using the complex coacervation process between gelatin and gum arabic. Although a very recurrent method, it is not trivial to establish a single coacervation methodology to encapsulate different compounds. The optimal synthesis conditions, that resulted in the best yield and encapsulation efficiency, are 1:1 (m/m) wall-to-core ratio, a temperature of 40°C and agitation speed of 10,000 rpm. Optical microscopy analysis revealed that the microcapsules are spherical, have average diameters of 112 μm (*A. vera*) and 118 μm (microalgae) and do not form agglomerates. The microcapsules were characterized by the osmotic pressure at which they ruptured, allowing the calculation of their mechanical resistance, which resulted in 392 MPa (*A. vera*) and 425 MPa (microalgae). The presented optimized methodology to encapsulate both compounds aims to contribute to their efficient and rational use, especially in cosmeceutical applications.

Keywords: *cosmeceutics, gelatin, gum arabic, morphology, osmotic pressure.*

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1. Introduction

Microencapsulation is a promising technology that encapsulates active agents in a protective boundary, thus increasing the stability of the encapsulated material and extending its lifetime. Microencapsulation inhibits chemical reactions between the core material and external species, such as moisture, oxygen, and protects the encapsulated molecules against temperature variations, and other environmental stresses[1,2]. Microcapsules have several applications, for example, in smart coatings (self-healing coatings), and also in pharmaceuticals, food, textiles, and cosmetics[1,2]. Nowadays, there are several encapsulation methods. Among the techniques used for the microencapsulation of active components, complex coacervation, which is considered the oldest method, stands out as a fast and simple method. The coacervation technique involves the interaction between hydrocolloids of opposite charges. Driven by the neutralization of the charges, the polyelectrolytes undergo phase separation. Thus, a two-phase system, comprising of a colloid-rich phase and a colloid-poor phase is formed. The colloid-rich phase coalesces around the dispersed material (an oil, for example), thus forming the microcapsule[3,4]. Complex coacervation allows the production of microcapsules using charged biomacromolecules with high efficiency, as well as the encapsulation of hydrophobic materials at mild temperatures[3,5-7].

The complex coacervation method has recurrently being used in food and cosmetics industries, to encapsulate a diversity of materials. Gelatin/gum arabic microcapsules, containing broccoli particles, were prepared by complex coacervation; aiming to preserve the encapsulated components, increase the chemical stability, and mask the aroma[8]. Peppermint oil and poppy seed oil were also encapsulated by complex coacervation of gelatin/gum arabic[7,9]. Nevertheless, not all materials are prone to be encapsulated, therefore the establishment of proper methods is required. Also, from the industrial perspective, the development of methods that can be applied for the encapsulation of different compounds is advantageous from the economical point of view.

In the cosmetic industry, cosmeceuticals are gaining much attention since they are an alternative to deliver active ingredient in cosmetics formulations[10]. In particular, some oils that have antioxidant, bactericidal, anti-inflammatory, and analgesic properties, have been the focus of various studies. For example, *Aloe vera*[11] is a plant of African

origin that was used for many years in traditional medicine. It is known for its foliage, which contains a mucilage-like gel. *A. vera* oil is obtained from the gel, which contains carbohydrates (glucomannan and acemannan), mineral salts (calcium, sodium, zinc, magnesium, chrome and copper) and vitamins (A, B12, C, E, choline and folic acid). When applied topically to the skin, the oil has beneficial properties preventing scaring and also having anti-inflammatory, hydrating, bactericidal, antiviral, and antifungal effects[11-14].

Another interesting source of compounds of cosmeceutical interest are microalgae, which are photosynthetic organisms whose size can vary between 5 and 50 μm and are found in both fresh and salt water. Due to their high nutritional value, they have been prominent in the economic area besides their ecological benefit[15]. Among the various species of microalgae, *Acutodesmus obliquus*, also known as 'Curitibana' because of its origin in Curitiba (state of Paraná, southern Brazil), is known to adapt rapidly to different climatic conditions, for example, the absence of sunlight and average temperatures of 17 C. *A. obliquus* contains proteins, pigments (chlorophyll, carotenoids), carbohydrates (β 1,3 – glucan, alginates, carrageenans, cellulose), and lipids, whose content is particularly high[16]. Microalgae pigments have been used in the pharmaceutical and cosmetic industries because of their antioxidant properties in addition to their use as dyes in paper, food and textile industries[17]. The proteins present in microalgae have their most common application in food and nutritional supplementation because they have antioxidant, nutritive and immunostimulatory properties[15,17]. In addition, the lipids in their composition are rich in fatty acids of high commercial value. In this way, researches have been seeking different applications for microalgae and its derivatives, covering areas such as food, cosmetics, medicinal and agriculture[16,18-20].

To the best of our knowledge, there is only one study about the encapsulation of *A. vera* and none aiming the encapsulation of *A. obliquus* fatty acids using the complex coacervation method. Considering the promising application of both compounds and the simplicity and economic feasibility of the coacervation method, this study sought the best experimental conditions to encapsulate *A. vera* or *A. obliquus* fatty acids using gelatin and gum arabic as the constituent wall polymers. The studied variables were the wall-to-core ratio, temperature and agitation speed. The best experimental condition was selected according to the polymers yield and encapsulation efficiency. The capsules obtained at the best condition were evaluated with respect to their size, morphology and mechanical resistance. In addition, a detailed discussion is presented about the parameters and mechanisms involved in the coacervation process, thus this study contributes to the better performance of encapsulation processes from both the economic and efficiency points of view.

2. Materials and Methods

2.1 Materials

Commercial gum arabic (GA), type-A gelatin, and glutaraldehyde were purchased from Sigma–Aldrich. GA and type-A gelatin were previously characterized as described elsewhere[5]. *A. vera* oil was purchased from Brazilian industry, and fatty acids from *A. obliquus* were donated by NPDEAS (Núcleo de Pesquisa e Desenvolvimento de Energia Autossustentável – UFPR, Brazil). All other chemicals and reagents were of analytical grade.

2.2 Study of the complex coacervation experimental parameters

The most relevant experimental parameters for the complex coacervation method are temperature, concentration, agitation speed and pH. Regarding pH, it was previously demonstrated that the optimal pH for the coacervation of the studied materials is 4.00, so this was the adopted value[2,5]. In this way, the studied variables were wall-to-core ration (m/m) (1:1 or 1:2), temperature (40°C or 60°C) and agitation speed (900 or 10,000 rpm). *A. vera* oil was employed as the model core material.

Microparticles were produced as described in the literature[5]. Briefly, GA (2.5 wt%) was dissolved in distilled water with magnetic stirring at room temperature (25°C) overnight with continuous stirring. Gelatin (2.5 wt%) was dissolved in distilled water at 40°C with continuous stirring. The pH of each solution was measured to check that a pH of around 5.6 was achieved. The pH was adjusted with 0.1 mol L^{-1} sodium hydroxide solution when necessary. The GA and gelatin solutions were mixed at a selected mass ratio, and a certain amount of core material was added. Then, the resulting dispersion was stirred for 5 min at 40°C or 60°C. Subsequently, 80 mL of deionized water was added (at the same temperature), and the solution was stirred for 5 min. The pH was adjusted to 4.00 by adding HCl (0.1 mol L^{-1}), and the resulting suspension was slowly cooled to 10°C in an ice bath. Finally, the dispersions were transferred to a refrigerator and left for 4 hours for complete precipitation and storage before further use. The microcapsules were crosslinked using 1 mmol L^{-1} of glutaraldehyde for each gram of protein. The reaction was carried out at 25°C for 12 hours under magnetic stirring. The microcapsules were washed three times with distilled water and acetone to remove some residual core material. The drying process was performed under reduced pressure.

2.3 Encapsulation efficiency and yield percentage

The encapsulation efficiency (*EE%*) was determined by the quantitative determination of the encapsulated material, based on the procedure described in the literature[21]. A known mass of the synthesized dried microcapsules was macerated for 15 minutes, washed with acetone, and dried at room temperature. The resulting mass indicates the mass of the coating material of the capsules. Thus, it is possible to calculate the amount of encapsulated core material extracted with acetone.

$$EE\% = \left(\frac{extracted\ oil}{total\ oil} \right) x100 \quad (1)$$

To calculate the yield percentage (*Y%*), the dried microcapsules were weighed, and the mass ratio was calculated with respect to the initial total mass, as shown in Equation 2.

$$Y\% = \frac{mass\ of\ dry\ microcapsules}{total\ initial\ mass} x100 \quad (2)$$

2.4 Characterization of the microcapsules

The morphology of the microcapsules was observed by optical microscopy (Alphaphot YS2, Nikon). The diameter and size distribution of microcapsules were obtained by laser

granulometry using a Microprocessor S3500 Bluewave on a wet sample.

The chemical characterization was carried out using a Fourier transform infrared (FTIR) Vertex 70 spectrometer with HTS-XT microplate extension (Bruker OPTIK GmbH). Attenuated total reflectance and transmission modes with a diamond crystal were used in the observation range of 4000 to 400 cm^{-1} with a resolution of 4 cm^{-1}.

The net surface electric charge and stability of the microcapsules, in water, were analyzed using a zeta-potential analyzer (Brookhaven Zeta PALS). The electrophoretic mobility was converted into zeta potential values based on the Smoluchowski model. The measurements were performed in triplicate at 25°C.

2.5 Mechanical resistance of the microcapsules

The microcapsules dispersions were mixed with an equal volume amount of glucose solution, prepared at different concentrations, and left stirring for 1 hour. Optical microscopy (Alphaphot YS2, Nikon) was employed to determine the number of deformed capsules. In this calculation, at least 100 capsules were counted at each glucose concentration. The minimum concentration necessary to induce the deformation of 50% of the microcapsules was defined as the critical glucose concentration and was used to determine the critical osmotic pressure based on the calibration curve available in the literature[22]. The modulus of elasticity of the polymer wall was calculated using Equation 3.

$$\pi_c = 4\mu\frac{\delta}{R^2} \quad (3)$$

Here, π_c is the critical osmotic pressure (Pa), μ is the modulus of elasticity (Pa), δ thickness of the microcapsule wall (m), and R the radius of the capsule (m). The mean wall thickness values were obtained from the optical micrographs and scanning electron microscopy (SEM) images. SEM images were taken of cross-sectioned dried microcapsules, and thickness measurements were made using *ImageJ*. The radius of the microcapsule was calculated from the average diameters obtained.

3. Results and Discussion

3.1 Determination of the best synthesis parameters

The formation of polyelectrolyte complexes occurs upon the electrostatic interactions, which depend on the degree of ionization of the polymers that is determined according to the environmental pH[5]. The pH adjustment is a fundamental step that promotes the equilibrium of the charges of the polymers present in the medium leading to the interaction between the polymers allowing the formation of the coacervates. Also, the precise selection of the pH is important to achieve the maximum coacervation yield. To confirm that the ideal pH for the formation of the polysaccharide/protein complex found is the same as indicated in the literature[5] and to evaluate the behavior of the wall materials, the zeta potential of the solutions of starting polymers was determined as a function of pH, as shown in Figure 1.

As shown in Figure 1, the polyelectrolytic complexation between gum arabic and gelatin is favorable because gum arabic has negatively charged surface for the whole studied pH range. Thus, the deprotonation of the carboxylic groups in gum arabic yields an anionic polyelectrolyte that will interact with positively charged species. Gelatin is an amphoteric macromolecule; that is, it has basic and acidic functional groups and, thus, an isoelectric point. Based on the graph, gelatin has zero zeta potential at pH 5.8. This result evidences that the acidic and basic groups of gelatin are equivalently ionized at this pH, indicating charge neutralization, thus giving the isoelectric point. Because the encapsulation efficiency depends on the interaction of oppositely charged species, the optimal complexation condition is when both concentrations are concomitantly maximized[5,23]. As depicted in Figure 1, at pH 4.0, gelatin and gum arabic have the greatest difference between their surface charges, therefore indicating that the ideal interaction condition is at this pH. This result is in perfect agreement with previously reported results[5].

As already explained, coacervation is based on a complex mechanism since it involves variables such as wall-to-core mass ratio, temperature, agitation speed and pH adjustment. At the selected pH 4.0, different synthesis variables were employed and the results in terms of the yield percentage (*Y%*) and encapsulation efficiency (*EE%*) are displayed in Table 1.

Based on Table 1, it was found that samples 2, 3, 4, 7 and 8 had yields above 80%. Analyzing the studied variables, it was observed that the best yield results were for the samples performed at 40°C, thus indicating that the temperature influences the phase separation (coacervation). Since high temperatures promote the increase of the diffusion coefficient of the macromolecules and their internal energy, the formed complexes can be destabilized, thus reducing the yield. Conversely, low temperatures favor the hydrogen bonds, aiding in the formation of stable polyelectrolyte complexes[3].

Samples 1, 3, 5 and 7, both synthesized at an agitation speed of 900 rpm, exhibited a greater amount of free oil at the end of the coacervation process. Among them, samples 5 and 7 showed the highest amount of free residual oil, since for these samples the wall-to-core ratio was 1: 2. These results corroborate with the values of the encapsulation efficiency, demonstrating that at 1:2 not enough material is available to form the capsule's wall, thereby resulting in a higher concentration of free core material in the medium, thus reducing the encapsulation efficiency[24].

The agitation speed is a parameter which influences, in particular, the characteristic of the formed emulsion. Table 1 shows that samples 2, 4, 6 and 8, in which the stirring speed of 10,000 rpm was used, when compared to the samples obtained at a speed of 900 rpm, exhibits a difference in both

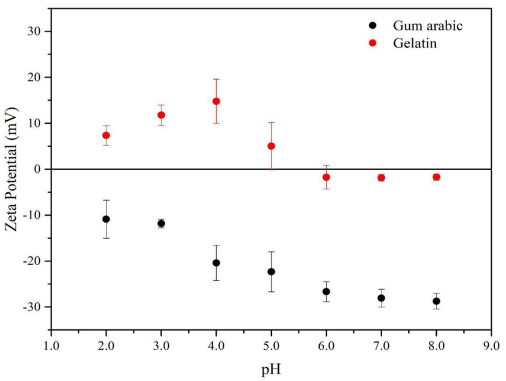

Figure 1. Zeta potential (mV) of the initial biopolymer's dispersions, gelatin and gum arabic, both at 2.5 wt%.

encapsulation efficiency and yield. This is because lower agitation values (<2,000 rpm) make it difficult to break the droplets of the material to be encapsulated and to form a stable emulsion [25].

In this way, it was possible to verify that sample 4 displayed the better results, since it presented an encapsulation efficiency of 79.6% and a yield of 89.8%. Therefore, the condition selected as the most favorable to achieve the higher yielding and encapsulation efficiency was 1:1 (m/m) wall-to-core ratio, 40°C and 10,000 rpm.

3.2. Preparation of the microcapsules containing A. vera and microalgae fatty acids

Considering an oil-in-water (o/w) emulsion, the high interfacial energy between water and oil is the driving force that promotes the formation of the coacervates around the oil droplets, thus reduction of the total energy of the system. For this to occur, the coacervates must exhibit a hydrophobic region, which will interact with the microcapsule nucleus (oil), and a hydrophilic region that will interact mainly with the solvent (water). When the core material is highly hydrophobic, it is necessary to use surfactants which will increase the affinity of the core with the shell, enhancing the encapsulation efficiency.

The *A. vera* oil contains several active components, among them the fatty acids (salicylic acid, gamma-linolenic acid, arachidonic acid and cholesterol). Equally, *A. obliquus* fatty acids are described as containing arachidonic acid, linoleic, oleic and alpha linoleic acids. Due to their chemical composition, these compounds exhibit amphiphilic character[14,20]. Therefore, both compounds were successfully encapsulated by the gelatin-gum arabic complex without the need to use surfactants.

Figure 2 shows photomicrographs of the gelatin/gum arabic microcapsules filled with microalgae fatty acids and *A. vera* oil. As observed, microcapsules containing both

Table 1. Experimental parameters of the coacervation between gelatin (2.5 wt%) and gum arabic (2.5 wt%), using calendula oils as the core material.

Sample	Wall-to-core mass ratio	Temperature (°C)	Speed (rpm)	*Y%*	*EE%*
1	1:1	60	900	72.0 ± 1.0*	28.7 ± 1.6*
2	1:1	60	10,000	81.6 ± 0.7*	43.4 ± 1.2*
3	1:1	40	900	83.0 ± 2.0*	34.4 ± 1.0*
4	1:1	40	10,000	89.8 ± 0.5*	79.6 ± 0.6*
5	1:2	60	900	69.6 ± 0.7*	36.4 ± 0.7*
6	1:2	60	10,000	78.4 ± 1.3*	58.6 ± 1.0*
7	1:2	40	900	84.9 ± 1.2*	41.3 ± 0.9*
8	1:2	40	10,000	85.3 ± 1.0*	50.7 ± 1.3*

*Average values determined in triplicate.

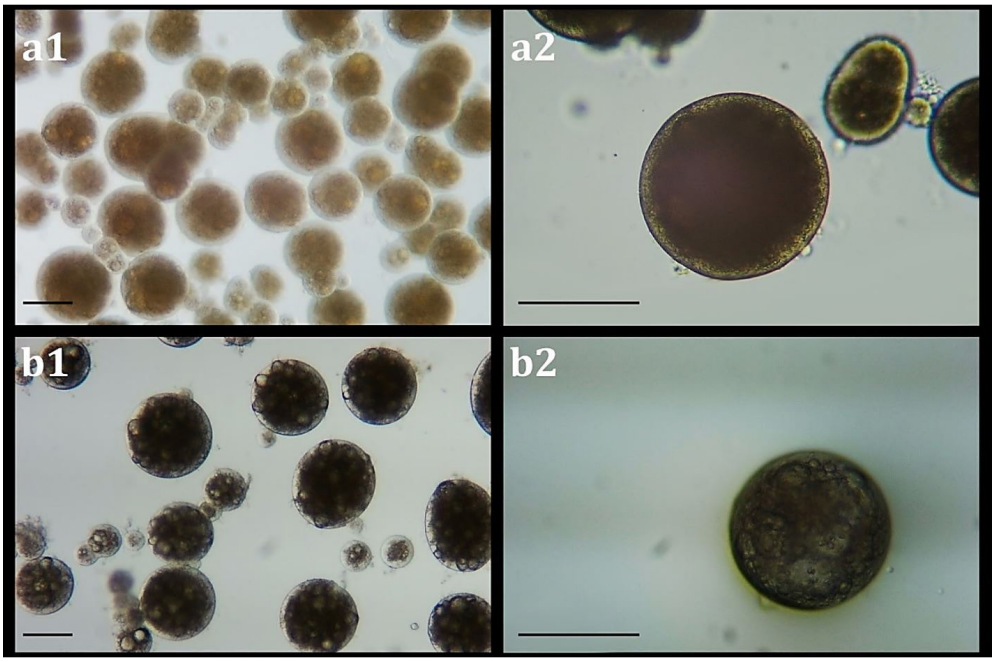

Figure 2. Optical microscopy images of synthesized microcapsules: (a1) gelatin/gum arabic microcapsules containing fatty acids from microalgae 40X and (a2) 100X; (b1) gelatin/gum arabic microcapsules containing *A. vera* oil 40X and (b2) 100X. Scale bars = 100 µm.

compounds could be successfully synthesized by the complex coacervation technique. All microcapsules have a spherical morphology with well-defined shells and a multinuclear structure. Spherical particles are desirable because they exhibit higher fluidity and a lower surface/volume ratio, allowing the complete coverage of the core material, thus favoring material retention[26]. The formation of continuous shells is very important for the microcapsules, ensuring material retention and the protection of the encapsulated material. During the observations, the microcapsules remained stable, and no rupturing was observed.

Figure 3 shows the SEM image of the cross section of a microcapsule containing *A. vera* oil. Based on this image and on the optical microscopy images and using *ImageJ*, the calculus of the average shell thickness is 3.5 µm.

The particle size (diameter) distributions for the capsules containing fatty acids and those containing *A. vera* oil are displayed in Figure 4.

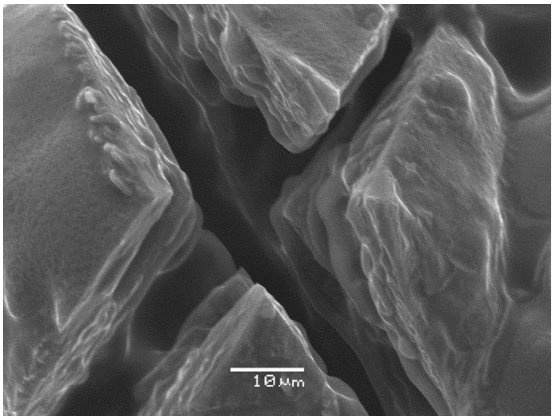

Figure 3. SEM image of a ruptured microcapsule containing *A. vera* oil showing the wall thickness. Scale bar: 10 µm.

As shown in Figure 4, the average diameter of the microcapsules obtained were 118 µm for microalgae fatty acids and 112 µm for *A. vera* oil and both types of microcapsules had a broad particle size distribution, ranging from 37 to 275 µm. The size of the microparticles determines their application, since larger capsules contain larger amounts of encapsulated material.

Furthermore, the yields obtained for *A vera* and *A. obliquus* microcapsules were all higher than 80% (80.2 ± 0.7 and 88.6 ± 0.3 for *A. obliquus* and *A. vera*, respectively) and the efficiencies of encapsulation were 78.3% (*A. vera*) and 77.9% (*A. obliquus*), indicating that the optimized conditions successfully produced microcapsules.

3.3 Chemical composition of the microcapsules containing A. vera and microalgae fatty acids

The FTIR spectra of gelatin (Gel), gum arabic (GA), microalgae fatty acids, *A. vera* oil, empty microcapsules (MC) and microcapsules filled with the materials are shown in Figure 5.

In the coacervation process, interactions between the positive and negative charges of the amino and carboxylic acid groups of the coating materials are expected. Based on the FTIR spectra, we verified that the gelatin/gum arabic wall of the capsule was formed because characteristics bands of gelatin and gum arabic are present in the spectrum of the empty microcapsules. In contrast, in the spectrum of the filled microcapsules, characteristic bands of the organic compounds present as the core materials are observed. Based on the spectra of the synthesized microcapsules, characteristic absorption bands are identified indicating contribution of both the wall and core materials. The broad band around 3290 cm^{-1} refers to NH and OH groups of amide A and to the OH groups of the gum backbone. The bands at 2923 cm^{-1} and 2853 cm^{-1} are relative to the stretching of CH bonds. The presence of carbonyl (C=O) groups of carboxylic acid

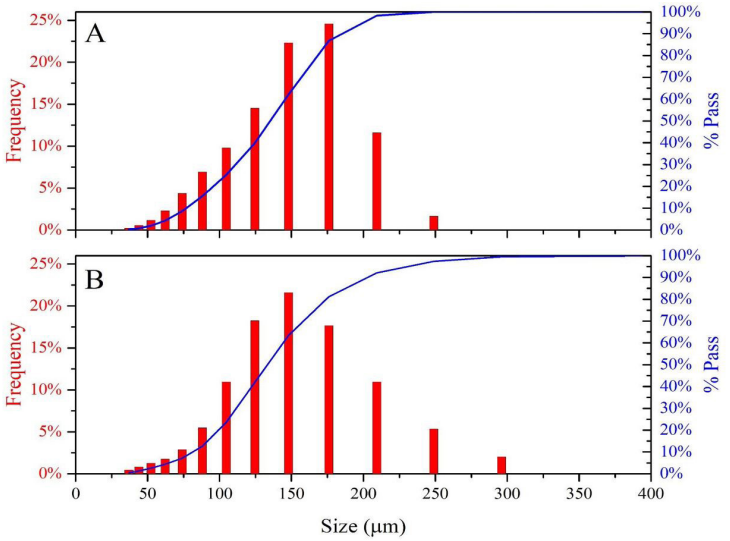

Figure 4. Particle size distribution obtained by laser granulometry: (a) microcapsules containing fatty acids from microalgae and (b) microcapsules containing *A. vera* oil.

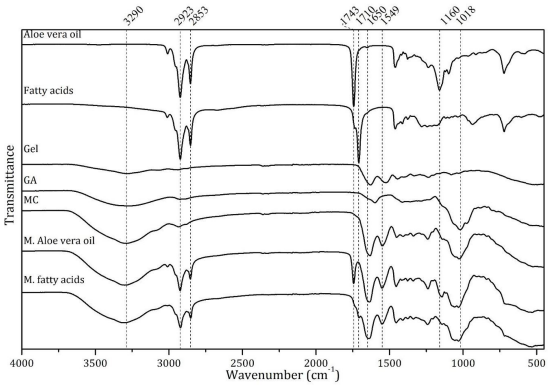

Figure 5. FTIR spectra of the core materials (Aloe vera oil and fatty acids), the shell polymers (Gel and GA), empty microcapsules (MC), and filled microcapsules with the respective material (M. Aloe vera oil and M. fatty acids).

Table 2. Materials characteristic bands used in the microcapsule's formation[7,27].

	Component	Band (cm^{-1})	Assignment
Shell materials	Arabic gum	3290	O-H stretching
		2923	C-H stretching
		1650	COO- symmetric stretching
		1018	C-O carbohydrates
	Gelatin	3290	N-H stretching
		2923	C-H asymmetric stretching
		1650	C=O stretching
		1549	N-H bend coupled with C-N stretching
Core materials	Microalgae fatty acids and *A. vera* oil	2923	=C-H stretching
		2853	-CH2 asymmetric stretching
		1743	C=O stretching
		1710	C=O stretching
		1160	C-O group stretching

(gum) and of amide I (gelatin) are evident at 1743 cm^{-1} and 1650 cm^{-1}, respectively. The stretching vibration of the C-O bond of esters present in the core materials can be seen at 1160 cm^{-1} and the band at 1018 cm^{-1} is consistent with the vibration of C-O groups in the gum.

It can be assumed that the desired compounds were effectively encapsulated in the gelatin/gum arabic microcapsules because the spectra of the empty microcapsules, the pure materials for encapsulation, and the filled microcapsules show significant similarities. Furthermore, we determined that there were no interactions between the core materials because no changes in the characteristic bands were observed when they were encapsulated. Thus, the complex coacervation process can be considered efficient for the synthesis and preparation of these microcapsules. The main characteristic IR bands of the core and shell materials are listed in Table 2.

3.4 Zeta potential determination and stability considerations

Most of the particulate materials acquire a surface electric charge when they are dispersed in solution, forming an electric layer on the surface of the particle. The first charged layer is called the Stern layer and can be charged positively or negatively depending on the characteristics of the particle and the medium. The next charged layer, which is more external than the Stern layer, is the diffusion layer, where the counter ions are strongly contributing to the total surface net charge. Since the particles diffuse in the medium along with the ions that are strongly attached to their surface, a shear plane can be defined as the location where the electric potential reduces exponentially as a function of the distance from the Stern layer. The value of the electric potential in the shear plane is called Zeta Potential (ζ) and is an indicative of colloidal stabilization[28].

In the case of capsules produced by polyelectrolytic complexation, where positive and negative charges interact by attractive forces, thus forming the capsule shell, the excess charge of this interaction results in the capsule charge. Thus, the zeta potential is an indicator of the electrostatic interactions that occurred between the polyelectrolyte molecules. The zeta potential values of the microcapsules of *A. vera* and microalgae fatty acids are -6.93 ± 0.69 mV and -6.51 ± 0.45 mV, respectively. Since the method formed particles with charges very close to zero, through neutralization of the constituent polymers, it is possible to infer that the coacervation process was very efficient and the mass ratio was properly selected. However, surface charges close to zero are very unfavorable from the stability point of view. Nevertheless, the microcapsules did not show aggregation and remained stable within the observation time of 15 days, then, the colloidal stability may be attributed to factors different from electrostatic repulsion. Sterically stabilized particles are an important class of polymer particles. When two microcapsules approach each other, the macromolecules meet at the particle interface. As a result, the interpenetration of these macromolecules occurs, and the degree of organization of the system increases, decreasing the entropy. Thus, the thermodynamics of aggregation is unfavorable, and particle separation is preferred, resulting in easy resuspension and preventing irreversible flocculation.

Concerning practical applications, because the obtained microcapsules have a slightly negative zeta potential, they could be applied in neutral to anionic cosmetic formulations without electrostatic interactions that would negatively affect the functionality of the final formulation.

3.5 Analysis of the mechanical properties of the microcapsules

The mechanical properties of the particles, such as rigidity, must be controlled for the processing of the final product. Thus, it was determined the mechanical properties of the capsules, specifically, the elastic modulus, also known as Young's modulus, which provides a measure of the stiffness of a solid material[29].

The micrographs of the capsules subjected to an osmotic pressure that caused their rupture are presented in Figure 6 and the values of the parameters related to the elastic modulus are shown in Table 3. After the minimum concentration of glucose necessary to promote the rupture of 50% of the microcapsules had been achieved, it was possible to calculate the elastic modulus of the polymer shell using Equation 3[30]. The obtained values of the elastic modulus

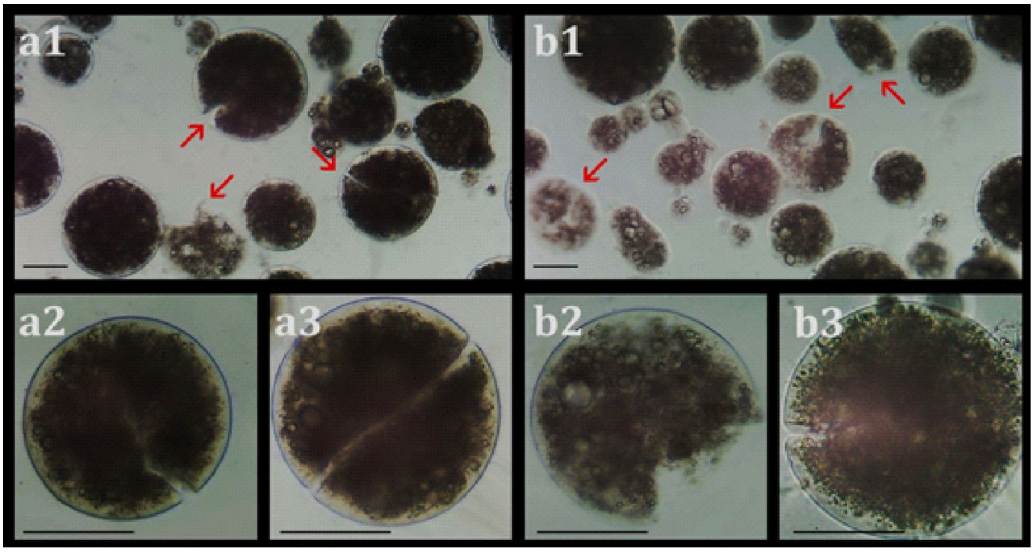

Figure 6. Optical microscopy images of gelatin/gum arabic microcapsules containing fatty acids from microalgae ((a1) 40X and (a2–a3) 100X) and microcapsules containing *A. vera* oil ((b1) 40X and (b2–b3) 100X) in 60% glucose solution for 1 h. Scale bars =100 μm.

Table 3. Glucose concentration, osmotic pressure, capsule thickness and radius, and percentage of capsules ruptured.

	Microalgae fatty acids			***Aloe vera* oil**		
Glucose concentration (%)	20	40	60	20	40	60
Osmotic pressure (MPa)	3.0	6.1	10.1	3.0	6.1	10.1
Shell thickness (μm)	4.5	4.5	4.5	4.5	4.5	4.5
Radius (μm)	59	59	59	56	56	56
Capsules rupture (%)	12	28	59	10	37	53

at the critical glucose concentration for microcapsules containing *A. vera* oil and for those containing microalgae fatty acids are 392 MPa and 425 MPa, respectively.

The chemical crosslinking following the synthesis of the gelatin and gum arabic microparticles by complex coacervation changes the release characteristics of the modified particles[31], as well as affects the strength of the capsules. Many crosslinking agents have reactive groups for protein immobilization, which can produce stable bonds with specific residues. The most common reagent for the formation of microcapsules using polysaccharides and/or proteins is glutaraldehyde because aldehydes react rapidly and form strong bonds with the polymeric microcapsule walls[31]. The crosslinking degree is proportional to the crosslinking reagent concentration. At low concentrations, intermolecular crosslinking is dominant. In contrast, at high crosslinking concentrations, more transverse intermolecular bonds are formed, resulting in low protein solubility[32]. It is known that crosslinked particles prepared with a high concentration of glutaraldehyde are more resistant to the spray drying process[2].

Considering that, in a typical process of packing, the material may be subjected to a pressure ranging from 0.065 MPa to 160 MPa[33], the magnitude of the elastic modulus found for the microcapsules prepared in this work demonstrates that they can be submitted to real conditions of transportation, packaging and storage without disruption. It is worth mentioning that the mechanical strength of the microcapsules can be properly varied according to the desired application by modifying the nature and concentration of the crosslinking agent.

4. Conclusions

Microencapsulation is a technology used to provide protection to the active agents so increasing their stability, performance and safe use. Nevertheless, not all agents are prone to be encapsulated. The complex coacervation process is a suitable method for the synthesis of microcapsules of gelatin / gum arabic containing microalgae fatty acids or *A. vera* oil. In this study, the best parameters for the complex coacervation method was determined, being obtained yields > 80% and encapsulation efficiency > 77%. The microcapsules were found to be spherical and multinuclear. The chemical analysis confirmed the formation of the polymeric microcapsule wall and the encapsulation of the core material, which did not evidence interactions between the wall and the core materials. The synthesized microcapsules are sterically stabilized and have a slightly negative surface charge. The mean particle diameter was 112 μm and 118 μm (*A. vera* oil and microalgae fatty acid microcapsules, respectively), suggesting that the capsules may encapsulate a sufficient amount of oil for different applications. Therefore, the proposed method of encapsulation of *A. vera* oil and fatty acids from the "Curitibana" microalgae by complex coacervation is efficient and feasible for industrial use.

Finally, osmotic pressure tests showed that the synthesized capsules presented good mechanical resistance allowing a wide variety of possible applications. Thus, the crosslinking conditions used were appropriate for the capsules studied.

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