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Double emulsion stage prior to complex coacervation process for microencapsulation of sweetener sucralose

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ABSTRACT

Microencapsulation has proven viable for various industrial applications. In the case of sweeteners, microencapsulation can increase the fluidity and resistance to high temperatures and prolong sensation of sweetness. The aim of this study was to microencapsulate sucralose by double emulsion followed by complex coacervation. The microcapsules were evaluated by optical and scanning electron microscopy, hygroscopicity, solubility, moisture, water activity, particle size, encapsulation yield, potential ZETA, fourier transform infrared spectroscopy (FTIR) and thermal behavior. The microcapsules presented low hygroscopicity and solubility, and average size ranging from 81.04 to 113.49 μm. With FTIR, it was possible to observe the amide bond that confirmed the formation of coacervates. Zeta potential showed that two samples presented neutral charge, indicating complete coacervation. The Tg values were above room temperature (53.59 to 56.88 °C). Among the formulation studied, the one produced with 5% gelatin and gum Arabic and core material 75% presented the best characteristics.

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1. Introduction

Sucralose is a synthetic sweetener derived from sucrose by selective replacement of three hydroxyl groups by chlorine atoms, resulting in substantial increase sweetness. It is soluble in water and ethanol and has sweetness intensity 400–1000 times sweeter than sucrose. The time-intensity profile is very similar to sucrose without bitter notes or metallic taste (Goldsmith and Merkel, 2001; Grenby, 1991; Hood and Campbell, 1990; Wallis, 1993).

Microencapsulation has shown great promise for incorporating some ingredients and additives in foods. In the case of sweeteners, this process is usually used with the purpose of increasing fluidity and resistance to high temperatures and extending the sensation of sweetness through the gradual and controlled release (Gouin, 2004).

The complex coacervation (CC) consists of a spontaneous separation of phases by forming a complex that can be insoluble, between two and more polymers resulting from electrostatic interactions (Yeo et al., 2005). However, this technique is suitable for encapsulating lipophilic materials. Since sucralose is a hydrophilic compound, to enable the use of this technique an adaptation was proposed in this study. Thus, a primary emulsion W/O was performed prior to CC, followed by double emulsion W/O/W.

Microcapsules produced by CC are water insoluble, temperature resistant and have excellent characteristics for controlled release (Dong et al., 2011). These characteristics are fundamental to achieve the aim of this study, which is to develop a vehicle for gradual release of sucralose during chewing.

No scientific paper discloses a microencapsulation method of sweeteners in the literature, but this topic has attracted great interest since there are many patents involving this matter. Given the above, the aim of this study was to encapsulate sucralose using a double emulsion technique followed by CC and characterize the microcapsules obtained.

2. Materials and methods

2.1. Materials

Sucralose sweetener (Techno Food Ingredients Co – CA, USA) was used as the core material. Bovine gelatin (GE) (Gelita – Cotia/SP, Brasil), and gum Arabic (GA) (Synth Diadema/SP, São Paulo, Brasil) were used as encapsulating agents. Lecithin (Gerbras Química Farmacêutica Ltda.) was used as emulsifier and soybean oil (Bunge-São Paulo/SP, Brasil) was used to produce the primary emulsion.

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2.2. Production of microcapsules by complex coacervation

The methodology for production of microcapsules by complex coacervation (CC) used in this study was previously described by Mendanha et al. (2009). An emulsion was prepared with 30% aqueous sucralose solution, soybean oil (twice the volume of sucralose solution) and soy lecithin (3% of total solids concentration basis). Then, the GE solution at 50 °C (at 2.5 or 5%) was added under constant stirring (12,000 rpm, 3 min) using Ultraturrax (IKA, T25, Germany). The same volume of GA solution (at 2.5 or 5%) and 4 times the volume of water were added to the emulsion, followed by adjusting the pH to 4.0 with HCl 0.1 M. The emulsion was cooled at 10 °C and stored at 7 °C for 24 h for complete separation of phases. After this period, the solutions were frozen for 24 h and freeze-dried. Six formulations with different concentrations of encapsulating agents (GE and GA) and different core material (sucralose in oil emulsion) as a function of the total content of encapsulating agents were obtained. The formulations were called as follows: A: 2.5% GE + GA and core material 50%; B: 2.5% GE + GA and core material 75%; C: 2.5% GE + GA and core material 100%; D: 5.0% GE + GA and core material 50%; E: 5.0% GE + GA and core material 75%; and F: 5.0% GE + GA and core material 100%.

2.3. Characterization of sucralose microcapsules

2.3.1. Morphological characteristics

The microcapsules were examined by optical microscopy (OM) and scanning electron microscopy (SEM). MO images were assessed by optical microscope (BEL photonics – Osasco/SP, Brasil) equipped with integrated 1.3 MP digital camera, and SEM images were assessed by scanning electron microscope (Hitachi TM-3000, Hiscope – New Jersey, USA), using 15 kV voltage.

2.3.2. Moisture content and water activity (Aw)

The moisture content of microcapsules and unencapsulated sucralose was determined by Ohaus MB-35 moisture analyzer balance, and water activity was performed on Aqualab water activity analyzer (Series 3 TE Decagon Devices-USA).

2.3.3. Solubility

The solubility was determined by gravimetric method, according to Eastman and Moore (1984), cited by Cano-Chauca et al. (2005). The sample (0.5 g) was added in an Erlenmeyer flask containing 50 ml of distilled water and the system was homogenized at 110 rpm for 30 min, followed by centrifugation at 4000 rpm for 5 min. An aliquot of 25 mL of supernatant was transferred to a porcelain dish of known weight and kept in oven at 105 °C to constant weight. The mass of empty dish and dish containing the dried material were taken into account for calculation the solubility of the microcapsules.

2.3.4. Hygroscopicity

About 0.5 g of sample was weighed in plastic dishes and stored for 7 days in a closed container containing saturated solution of anhydrous Na_2SO_4 (81% RH). The hygroscopicity was expressed as mass of water absorbed per 100 g of sample (Cai and Corke, 2000).

2.3.5. Particle size

The average particle size was assessed by a particle analyzer by laser diffraction (SALD – 201 V, Shimadzu -Japan) with a measurement range between 0.5 and 500 μm. The particles were dispersed in isopropanol and stabilized for 5 min before the analysis.

2.3.6. Zeta potential measurements

The Zeta potential measurements were performed for the microcapsules, encapsulating agents, and unencapsulated sucralose by a Zeta Potential Analyzer (BTC – Brookhaven Instrument Corporation – USA) in 10 runs of three cycles each, by diluting the samples in 1 mM KCl solution.

2.3.7. Fourier transform infrared spectroscopy (FTIR)

Analyses of sucralose, ingredients and microcapsules were obtained in the spectral wavelength range from 600 to 4000 cm^{-1}, by Perkin Elmer FT-IR Spectrometer with the aid of software Spectrum one v 5.3.1.

2.3.8. Encapsulation yield

The encapsulation yield (EY) was calculated according to Junxia et al. (2011) as the ratio of the total sweetener present in the capsule (E_{total}) and the amount of sweetener used to produce the microcapsules ($E_{production}$), as shown in Eq. (1).

To determine the total content of sweetener present in the microcapsules, 5 ml of 1% saline solution and 5 mL acetonitrile were added to falcon tubes containing 0.1 g of freeze-dried microcapsules. The tubes were shaken in a shaker tube and exposed to ultrasonic for 5 min, followed by centrifugation (4000 rpm) for 5 min. Then, an aliquot of the supernatant was removed for analysis, which was performed by external standardization in a liquid chromatograph (Shimadzu Prominence, Japan) equipped with a quaternary pump, auto injector (SIL – 10AF), reverse phase column (Shim-pack VP-ODS; 250 × 4.6 mm), diode array detector (210 nm) and data software (LC solution), according to the methodology described in IAL, 2005.

$$\text{EY}\ (\%) = \frac{E_{\text{total}}}{E_{\text{production}}} \times 100 \quad (1)$$

2.3.9. Differential scanning calorimetry

Samples (~1 g) were equilibrated in air over dry silica gel at 25 °C. After equilibration (about 2 weeks), aliquots were taken for DSC analysis. Phase transitions were determined by differential scanning calorimetry using a DSC TA2010 controlled by a TA5000 module (TA Instruments, Newcastle, USA). Samples of about 10 mg, conditioned at TA aluminum pans were heated between −20 and 200 °C at a rate of 10 °C/min, in N_2 inert atmosphere (45 mL/min). An empty pan was used as reference. Liquid nitrogen was used for sample cooling before the runs. Samples showing a devitrification peak after the first run were annealed at the devitrification peak temperature (Td) for 30 min before the second DSC run. Phase properties were determined by the DSC thermograms using the Software Universal Analysis V1.7F (TA Instruments, Newcastle, USA).

2.4. Statistical analysis

The data were evaluated by ANOVA ($p < 0.05$) and Tukey's comparison test ($p < 0.05$) using the Statistica software (Statsoft, USA).

3. Results and discussion

Fig. 1A shows one of the emulsions obtained after the addition of soybean oil in the aqueous solution of sucralose and lecithin. The presence of droplets and their movement (Brownian motion) on the microscope slide indicated the formation of the emulsion, which remained stable without phase separation for at least two hours. Fig. 1B presents the MO image of one of microcapsule formulations, which confirms that encapsulation of the sucralose was successful. All formulations exhibited similar OM and SEM

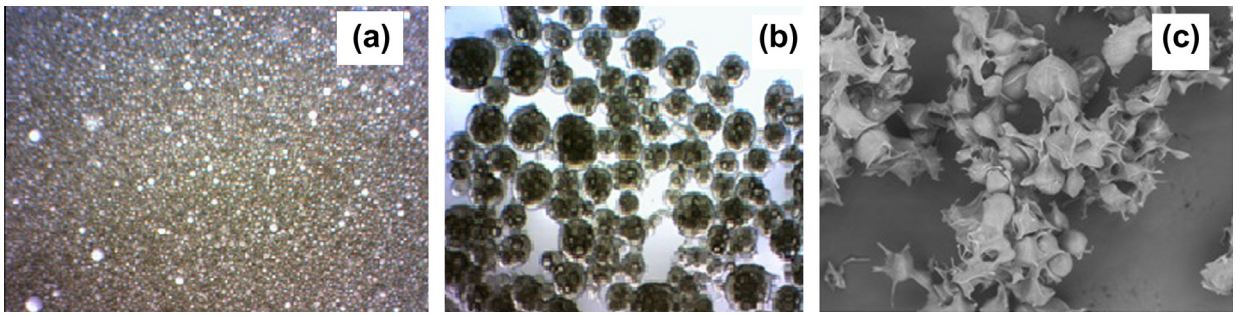

Fig. 1. Optical microscopy of formulation E (5% GE + GA and core material 75%) with 40 and 10× magnification (a and b) and scanning electron microscopy with 200× magnification (c).

images, with spherical shape and defined walls. Also in Fig. 1B, it can be seen that microcapsules are multinucleate and the sucralose emulsion droplets were distributed through the center of the capsules but not on its walls, like a reservoir system, which, according to Dong et al. (2011), gives excellent characteristics of controlled release, which is a major goal to encapsulate sweeteners.

In Fig. 1C is shown a SEM image of the dried capsules after the lyophilization process. The microcapsules had continuous walls with no cracks or apparent porosities, indicating that the lyophilization process was adequate, since it did not cause damage to the particles. These characteristics are important to ensure greater protection and retention of the encapsulated material. The capsules are cross-linked by solid bridges, which were also observed by Prata et al. (2008) in the encapsulation of vetiver oil by CC using GE and GA as encapsulating agents. These solid bridges can be attributed to the lyophilization process, which clusters the microcapsules. After rehydration, the lyophilized microcapsules exhibited the shape shown in Fig. 1B.

Table 1 shows the values obtained for particle size, moisture content, aw, solubility, hygroscopicity, Zeta potential, encapsulation yield and glass transition temperature.

The average particle size of the microcapsules ranged from 81 to 86 μm for formulations D, E and F (5% GE + GA) and from 100 to 113 μm for formulations A, B and C (2.5% GE + GA). As observed by Mendanha et al. (2009), varying the core material did not cause major changes in particle size, however it was observed that the concentration of encapsulating agents have influenced the particle size, since the mean size of the microcapsules of formulations A, B and C were higher as compared to other formulations. This influence is cited in several studies that associate the size of the microcapsules with production parameters such as ratio of polymer wall, concentration of polymers used, stirring speed, cooling rate, and drying process (Lamprecht et al., 2000; Menger et al., 2000; Nakagawa et al., 2004).

The moisture content and aw are within both the expected range for powder products and recommended to guarantee the microbiological stability. With respect to solubility, there was a significant reduction (4–10 times) as comparing the encapsulated samples with unencapsulated sucralose. The low solubility is a characteristic expected for the microcapsules obtained by CC and it is desirable in the encapsulation of sweeteners, since possibly contributes to the gradual release of the sweetener during chewing, prolonging sensation of sweetness.

The hygroscopicity values of the microcapsules ranged from 5.16 to 16.11 g water absorbed/100 g sample. These values were considered low, thus it may favor packaging and material handling. The values obtained in this study were lower (up to 6 times) than those determined by Nori et al. (2011) in propolis microcapsules obtained by CC using soy protein isolate and pectin as encapsulating agents.

With regard to the Zeta potential values, at pH 4, positive electric charges predominated in GE solution while negative electric charges predominated in GA, which is important to promote coacervation of these polymers. Concerning the microcapsules, samples E and F presented Zeta potential toward zero, which means that the charges were neutralized as expected in CC process. The remaining samples presented negative Zeta potential values, which indicate that negative charged groups remained, probably from GA, since GE is an amphoteric substance thus in acid medium positive charges may predominate. This result shows that these negative charged groups were not involved in electrostatic bonds to promote coacervation.

The EY ranged from 43.04 to 89.44%, thus it can be considered a good result concerning the hydrophilic material of this study, which has been previously emulsified in oil and encapsulated by CC. For both samples with 2.5% or 5.0% of encapsulating agents, the higher the core material, the lower the EY. This result may be due to the limited emulsifying capacity of GE in the interface O/

Table 1
Means and standard deviations obtained for the physicochemical properties of sucralose microcapsules, GE, GA, and unencapsulated sucralose.

Sample	PS (μm)	M (%)	Aw	S (%)	H (g/100 g)	ZP	EY (%)	Tg (°C)
A	113.49 ± 0.02	11.19 ± 2.35^{adg}	0.51 ± 0.18^{ae}	20.91 ± 1.60^{ae}	16.11 ± 2.78^{a}	-3.76 ± 6.48^{a}	89.44^{a}	53.59
B	100.15 ± 0.05	11.4 ± 1.19^{adg}	0.63 ± 0.05^{adg}	9.93 ± 0.93^{b}	9.98 ± 0.42^{c}	-4.16 ± 6.51^{a}	67.63^{b}	55.88
C	104.15 ± 0.05	6.68 ± 0.53^{bc}	0.35 ± 0.04^{ce}	9.66 ± 2.01^{b}	7.21 ± 0.40^{bd}	-18.78 ± 5.46^{b}	61.06^{c}	56.87
D	86.77 ± 0.08	13.97 ± 0.27^{d}	0.68 ± 0.06^{af}	14.17 ± 3.04^{bd}	7.19 ± 0.31^{bd}	-5.14 ± 8.52^{a}	68.56^{b}	53.75
E	81.04 ± 0.08	13.06 ± 1.54^{ad}	0.63 ± 0.06^{abh}	23.75 ± 2.01^{a}	6.71 ± 0.11^{cd}	0 ± 0^{a}	51.04^{d}	56.61
F	83.92 ± 0.08	12.79 ± 1.81^{ade}	0.77 ± 0.14^{bdf}	16.89 ± 2.09^{ed}	5.16 ± 0.26^{cd}	0 ± 0^{a}	43.04^{e}	56.88
GE	–	11.99 ± 0.40^{adf}	0.61 ± 0.02^{adg}	31.91 ± 1.71^{c}	27.16 ± 0.06^{e}	13.7 ± 0.97	–	73.87
GA	–	9.17 ± 0.32^{ecfg}	0.41 ± 0.01^{egh}	95.70 ± 0.62^{f}	38.13 ± 1.73^{f}	−21.33 ± 0.82	–	56.06
Suc	–	9.45 ± 1.41^{ac}	0.58 ± 0.05^{aed}	100.40 ± 0.79^{g}	0.08 ± 0.00^{g}	–	–	113.58

The same letters in the same column do not differ ($p < 0.05$). PS = particle size; M = moisture; Aw = water activity; S = solubility; H = hygroscopicity; ZP = zeta potencial; EY = encapsulation yield; Tg = glass transition temperature; Suc = sucralose. A: 2.5% GE + GA and core material 50%; B: 2.5% GE + GA and core material 75%; C: 2.5% GE + GA and core material 100%; D: 5.0% GE + GA and core material 50%; E: 5.0% GE + GA and core material 75%; and finally F: 5.0% GE + GA and core material 100%.

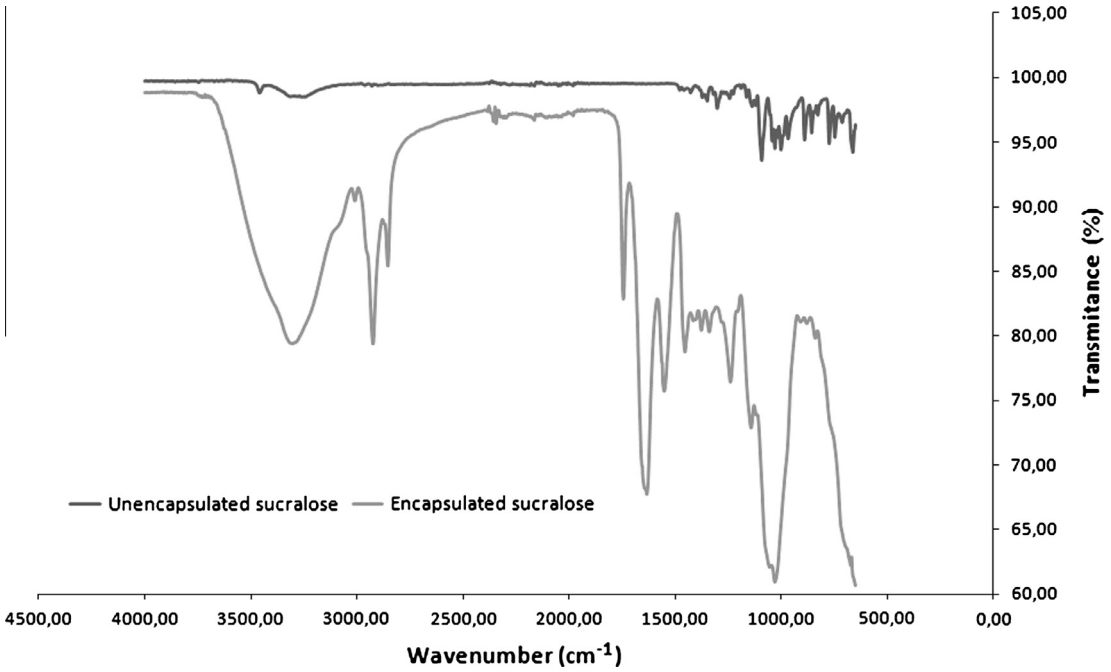

Fig. 2. FTIR of encapsulated and unencapsulated sucralose.

W in the presence of excessive concentration of primary emulsion (water in oil). Possibly, if less core material had been used the results for this parameter would have been even more satisfactory because GE would have acted more effectively in the interface between primary and secondary emulsion, avoiding the loss of core material. This result was similar to that reported by Jun-Xia et al. (2011), who encapsulated orange essential oil by CC using soy protein isolate and GA as encapsulating agents. These authors varied the core material from 10 to 70% as a function of the encapsulating agent and found the highest yield in the formulation with core material 10%.

Furthermore, similar core material showed slightly higher EY in the microcapsules produced with 2.5% encapsulating solution. According to Thies (1995), the increased viscosity caused by the high polymer concentration may interfere with capsules formation, since the mobility of the macromolecules may decrease and consequently increase the competition for solvent molecules.

The capsules showed similar and relatively high *Tg* values (53 to 56 °C). Obtaining *Tg* values above room temperature is a very positive result, as it ensures physical stability of the capsules when stored at room temperature, whereas, according to Roos (1995), physical changes such as agglomeration and collapse of the structure may occur when the *Tg* is below the storage temperature.

Fig. 2 shows the spectra of a formulation of encapsulated and unencapsulated sucralose and. GA is a polysaccharide having free carboxyl groups that confer negative charge to this molecule, while GE is a positively charged protein in acidic medium, due to amine groups. During CC, carboxyl groups from polysaccharides interact with amino groups of proteins to form a complex containing amide. FTIR analysis verified the formation of amides in the samples, confirming the occurrence of CC. In the spectra of encapsulated samples, elongated peaks appeared around 2900 cm^{-1}, which are characteristic of carboxyl groups. There were also peaks between the wavelengths 3400 and 3550 cm^{-1}, which are characteristic of amine groups. The presence of these peaks indicates that not all GA carboxyl groups interacted with GE amino groups, confirming the Zeta potential results. However, the presence of peaks characteristic of amides, which appear around 1500–1640 cm^{-1} confirms the coacervate formation.

According to the thermograms shown in Fig. 3, the microcapsules showed similar thermal behavior, as their thermograms did not differ. This result allows us to infer that changes in the core material and encapsulating materials did not alter the thermal behavior of the microcapsules. The microcapsules exhibited two melting peaks, at around 60 and 105 °C, which corresponded to melting peaks of gum Arabic and gelatin, respectively, as shown in the thermograms of these materials (Fig. 4). Sucralose showed a crystallization peak at about 115 °C and *Tg* at 113 °C, proving to be a material quite stable. Once no crystallization peak appeared in the thermograms of microcapsules, it suggested that sucralose remained in the amorphous form.

This study shows the viability of producing microcapsules of sucralose by double emulsion followed by CC. Concerning the six formulations studied, according to the particle size, it is suitable to use the formulations D, E or F for food applications, once

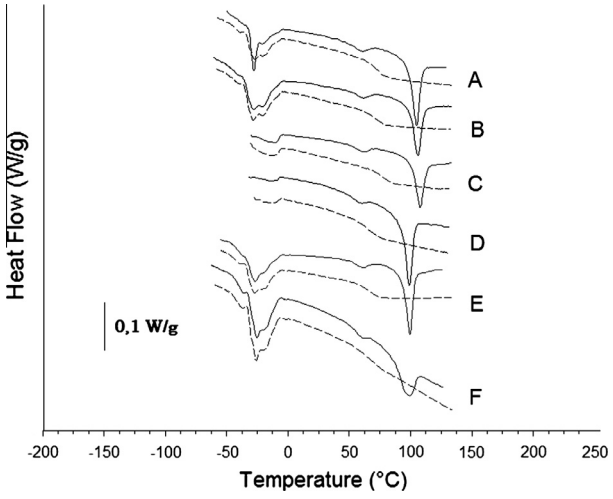

Fig. 3. Thermograms of the sucralose microcapsules. A: 2.5% GE + GA and core material 50%; B: 2.5% GE + GA and core material 75%; C: 2.5% GE + GA and core material 100%; D: 5.0% GE + GA and core material 50%; E: 5.0% GE + GA and core material 75%; and F: 5.0% GE + GA and core material 100%.

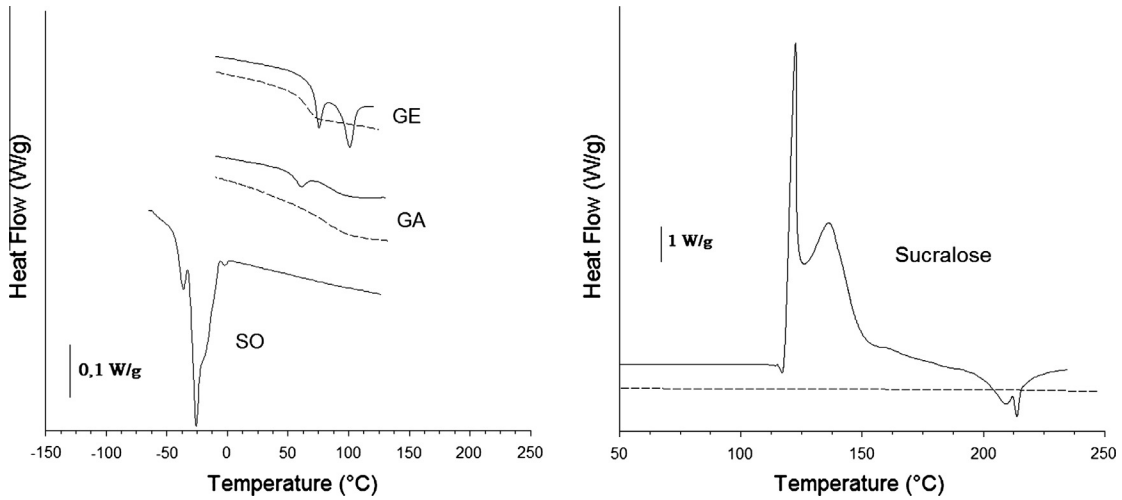

Fig. 4. Thermograms of the ingredients used to produce the microcapsules. GE = Gelatin, GA = gum Arabic and SO = soybean oil.

microcapsules are smaller than the others and can be easily incorporated into food products. Moreover, these microcapsules showed the lowest values for hygroscopicity, which facilitates transportation, storage and implementation. Regarding production parameters, formulation D showed higher EY than formulations E and F, so it may provide a better-controlled and gradual release of the sweetener, which is the main objective of this study. Therefore, formulation D is considered the best formulation for sucralose encapsulation as compared to the other formulations studied in the present paper.

4. Conclusion

Considering the aims and the results of this study, the proposed methodology of double emulsion followed by CC proved viable to encapsulate sucralose. Spherical and multinucleated microcapsules were formed, characteristics of complex coacervation technique, indicating that the double emulsion stage has been used successfully. The formulations have the potential to be applied in foods, especially formulation D, due to its most suitable characteristics. Future studies are required to the application of microcapsules in food products in order to study the release time and the effects on sensory characteristics of the products.

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