Food Chemistry 139 (2013) 72–78

Contents lists available at SciVerse ScienceDirect

Food Chemistry

journal homepage: www.elsevier.com/locate/foodchem

Microencapsulation of aspartame by double emulsion followed by complex coacervation to provide protection and prolong sweetness

Glaucia A. Rocha-Selmi [a,*], Fernanda T. Bozza [b], Marcelo Thomazini [b], Helena M.A. Bolini [a], Carmen S. Fávaro-Trindade [b]

[a] FEA, University of Campinas, Monteiro Lobato, 80, Campinas, Brazil
[b] FZEA, University of Sao Paulo, Duque de Caxias Norte, 225, Pirassununga, Brazil

ARTICLE INFO

Article history:
Received 26 November 2012
Received in revised form 16 January 2013
Accepted 23 January 2013
Available online 11 February 2013

Keywords:
Microcapsules
Controlled release
Sweetener
FTIR
Isotherm
Solubility

ABSTRACT

The objective of this work was to microencapsulate aspartame by double emulsion followed by complex coacervation, aiming to protect it and control its release. Six treatments were prepared using sunflower oil to prepare the primary emulsion and gelatin and gum Arabic as the wall materials. The microcapsules were evaluated structurally with respect to their sorption isotherms and release into water (36 °C and 80 °C). The microcapsules were multinucleated, not very water-soluble or hygroscopic and showed reduced rates of equilibrium moisture content and release at both temperatures. FTIR confirmed complexation between the wall materials and the intact nature of aspartame. The results indicated it was possible to encapsulate aspartame with the techniques employed and that these protected the sweetener even at 80 °C. The reduced solubility and low release rates indicated the enormous potential of the vehicle developed in controlling the release of the aspartame into the food, thus prolonging its sweetness.

1. Introduction

Aspartame is a dipeptide composed of L-aspartic acid and the methyl ester of phenylalanine, both amino acids found naturally in foods. It is about 160–220 times sweeter than sucrose and its flavour profile is described as clean and sweet like sucrose, without the bitter or metallic aftertastes normally associated with certain sweeteners such as acesulfame-K, cyclamate and saccharin (Butchko, Stargel, Comer, Mayhel, & Andress, 2001). It is inadequate for use in applications involving drastic heating for prolonged periods, such as baking, sterilisation and frying, since under such conditions, part of the molecule may undergo hydrolysis leading to a loss of sweet taste (Nabors, 2002; Salminen & Hallikainen, 2001).

The main objectives of microencapsulating sweeteners are to increase their fluidity and resistance to high temperatures and prolong the sensation of sweetness by controlling their release (Fávaro-Trindade, Pinho, & Rocha, 2008; Gouin, 2004). Thus this technology could facilitate the application of aspartame to products in which high processing temperatures are used, and also provide a gradual release of the sweetener when chewing, prolonging perception of the sweet taste of products such as sweets and chewing gum.

A good wall material should present the following characteristics: good emulsifying property and film-forming, low viscosity, low hygroscopicity, good drying properties, stability, absence of flavours and good protection to the core (Ré, 1998; Shahidi & Han, 1993). Gelatin, a water-soluble protein of high molecular weight and gum Arabic, a long chain polymer of high molecular weight is one of the most common and extensively utilised pairs in complex coacervation (Qv, Zeng, & Jiang, 2011). Microcapsules produced by complex coacervation are insoluble in water, resistant to high temperatures and show excellent characteristics for controlled release (Dong et al., 2011). These characteristics are desirable for the encapsulation of sweeteners, although the complex coacervation technique is appropriate for the encapsulation of hydrophobic compounds, which is not the case of aspartame. Mendanha et al. (2009) were successful in encapsulating a casein protein hydrolysate, which is also highly water-soluble, by adding a double-emulsion step at the start of the complex coacervation process.

A single paper was found in the literature whose objective was to study the stability of aspartame encapsulated in high melting point fat during the baking of cakes (Wetzel & Bellt, 1998). No other papers studying the microencapsulation of sweeteners were found in the literature, although it is a subject that stimulates great interest in industry, since there are numerous patents involving this subject. Thus the present paper could provide an impulse for the divulgation of new research on the technique of microencapsulating sweeteners.

* Corresponding author. Tel.: +55 1935214084; fax: +55 1935214060.
E-mail address: glaucia84@gmail.com (G.A. Rocha-Selmi).

0308-8146
http://dx.doi.org/10.1016/j.foodchem.2013.01.114

Hence the objective of the present work was to study the microencapsulation of aspartame by double emulsion followed by complex coacervation, structurally evaluate the microcapsules obtained, and also examine their physicochemical properties and rate of release into water.

2. Material and methods

2.1. Materials

The sweetener aspartame (AS) (Ajinomoto do Brasil, Brazil) was used as the core material, and swine gelatin (GE) (Gelita, Brazil) and gum Arabic (GA) (Synth, Brazil) as the wall materials. Sunflower oil (Cargill, Brazil) and soybean lecithin (Caramuru, Brazil) were used to prepare the primary emulsion.

2.2. Preparation of the coacervated microcapsules

The methodology used to prepare the microcapsules was an adaptation of that of Mendanha et al. (2009), who encapsulated a protein hydrolysate by complex coacervation, adding a double emulsion step at the start of the process. Three concentrations of AS solution were prepared based on preliminary tests: 3.75, 5 and 10 g/100 g of total solution, and were then emulsified at 50 °C with soybean oil in an Ultraturrax homogenizer (Turratec, TE-102, Tecnal, Brazil) for 3 min at 18 000 rpm, using soybean lecithin as the emulsifier (5 g/100 g of the total amount of AS). The ratios of sweetener:oil were 1:1, 1:2 and 1:3. The primary emulsion was emulsified in GE, and the GA solution subsequently added plus twice the total volume in distilled water. The pH value was adjusted to 4.0 and the system then cooled to 10 °C with constant stirring, and maintained in the refrigerator for 24 h to complete particle precipitation. The concentrations of the wall material were 2.5 and 5 g/100 g of total solution and the ratio of GE:GA maintained at 1:1 for all formulations. The amounts of core material were 50, 75 and 100 g/100 g of the total wall material used.

To increase the stability and facilitate handling, the coacervated microcapsules were freeze-dried. For this process, the excess water was removed and the coacervate frozen in a freezer at a temperature of −18 °C for 24 h, followed by freeze-drying in a bench-scale freeze dryer (Terroni LC 1500; São Carlos, Brazil).

2.3. Morphological evaluation

The morphology of the microcapsules was studied using an optical microscope (BEL Photonics®, Milan, Italy) equipped with a camera, using BEL View v. 62 software, and by scanning electronic microscopy (SEM) using the TM 3000 Tabletop Microscope (Hitachi, Tokyo, Japan) with the TM 3000 program. For SEM the microcapsules were placed on strips of double-faced carbon tape (Ted Pella, Inc., Redding, CA), which were then fixed to aluminium stubs. The images were captured with voltage acceleration of 5 kV and a current of 1750 mA.

2.4. Physicochemical characterisation of the coacervated microcapsules

2.4.1. Moisture content

The moisture contents of the freeze-dried material and of the non-encapsulated AS were determined automatically in moisture analysis equipment (MB45; Ohaus, Nänikon, Switzerland).

2.4.2. Solubility

A 1-g sample of each formulation was added to recipients containing 100 mL of distilled water, and stirred at 110 rpm for 30 min using a bench stirrer (Tecnal, Brazil), before centrifuging at 4000 rpm for 5 min. Aliquots of each supernatant were then removed with the aid of volumetric pipettes, transferred to previously weighed porcelain dishes, and dried to constant weight in an incubator at 105 °C. The dishes were weighed and the solubility calculated from the difference in weight (Cano-Chauca, Stringheta, Ramos, & Cal-Vidal, 2005).

2.4.3. Hygroscopicity

The analyses were carried out in triplicate, weighing approximately 1 g of each sample into circular plastic containers (diameter 40 mm × height 10 mm). The microcapsules were placed in hermetic pots containing a saturated sodium sulphate solution (relative humidity of 81%) and weighed again after 7 days. The hermetic pots were kept at 25 °C in an incubator with controlled temperature. The hygroscopicity was expressed as grams of water absorbed by 100 g of sample (Cai & Corke, 2000).

2.4.4. Particle size

The size and size distribution of the solid lipid particles were evaluated using a Sald-201V laser diffraction particle analyser (Shimadzu, Kyoto, Japan). The particles were dispersed in isopropanol (Synth, Brazil) and stabilised for 5 min before the analysis (Fávaro-Trindade, Santana, Monterrey-Quintero, Trindade, & Netto, 2010).

2.5. Encapsulation yield

The encapsulation yield (EY) was calculated according to Junxia, Hai-yan, and Jian (2011) as shown in equation 1. To determine the total AS present in the microcapsules, 0.1 g of freeze-dried microcapsules were weighed into a Falcon tube and 5 mL of a NaCl solution (1 g/100 g of solution) added plus 5 mL acetonitrile. The tubes were shaken in a tube shaker, placed in an ultrasonic bath for 5 min, centrifuged for 5 min at 4000 rpm and an aliquot of the supernatant removed for quantification of the AS.

The AS was quantified by external standardisation in a liquid chromatograph (Shimadzu), equipped with a spectrophotometric detector (214 nm), C18 column (250 mm; 4.6 μm) and a mobile phase of acetonitrile:methanol:phosphate buffer (pH 3.5) in proportions of 1:1:8, injecting a sample volume of 10 μL. The standard curve was prepared using concentrations of AS between 4.25 and 21.25 mg per 100 mL.

$$EY = \text{total AS}/\text{AS used in the production} \quad (1)$$

2.6. Fourier-transform infrared spectroscopy (FTIR)

The pure ingredients (gelatin, gum Arabic and aspartame) and the microcapsules (six formulations) were characterised by infrared spectroscopy in the region from 4000 to 600 cm^{-1}, using a Perkin Elmer FT-IR spectrometer (Waltham MA), with the aid of Spectrum One (version 5.3.1.) software.

2.7. Evaluation of the sorption isotherms of the powdered microcapsules

The sorption isotherms were determined by a gravimetric method. About 1 g of each sample, previously dried by exposure to P_2O_5, was weighed and placed in desiccators for 3 weeks at 25 °C, with relative humidities varying from 11% to 84%. The moisture content was calculated from the weight gain, and the experimental equilibrium moisture content obtained from the difference between the amount of water absorbed divided by the mass of the dry sample.

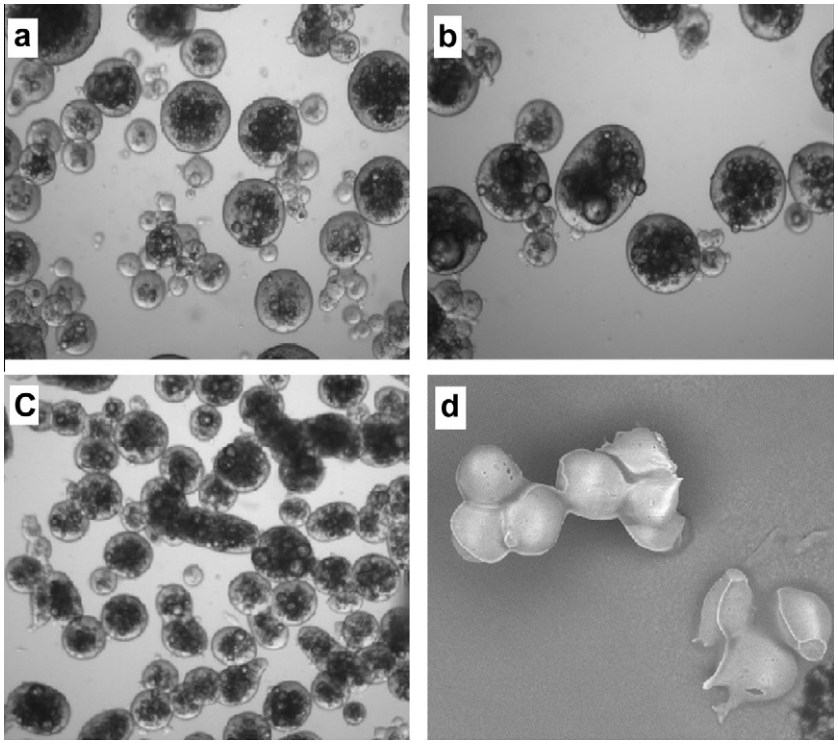

Fig. 1. Micrographs obtained with the optical microscope (a–c), and scanning electronic microscope with 500× magnification (d), of the aspartame microcapsules: (a and d) formulation A (2.5% GE and GA and 50% core material), (b) formulation B (2.5% GE and GA and 75% core material), (c) formulation C (2.5% GE and GA and 100% core material).

The sorption isotherms were better described by the GAB model (Eq. (2)), compared to BET and Peleg models.

$$Meq = (X_m CKa_w)/(1 - Ka_w)(1 - Ka_w + CKa_w) \quad (2)$$

where Meq – equilibrium moisture content (% dwb); a_w – water activity; X_m – moisture content of the molecular monolayer; C and K – parameters depending on the temperature and nature of the product.

2.8. Release of aspartame from the microcapsules into water at 36 and 80 °C

The release was analysed according to the methodology of Dong et al. (2011), with some modifications with respect to the equipment used. Falcon tubes containing suspensions with 5% (mass basis) of capsules were placed in a water bath with orbital shaking at temperatures of 36 (human body temperature) and 80 °C (to simulate heat treatment). Aliquots were removed after 0, 20, 40, 60, 80 and 100 min, for quantification of the AS using the same methodology used to determine the encapsulation yield.

2.9. Statistical analysis

The experiments were carried out in triplicates. Differences between mean values were determined using analysis of variance (ANOVA), utilising the statistical software SAS version 8.0 (SAS Institute Inc., Cary, NC).

3. Results and discussion

3.1. Preparation of the microcapsules and their morphology

Numerous preliminary trials were carried out, in order to define the concentrations of the ingredients and conditions for the production of the emulsions. Of the different emulsion formulations tested, those prepared with a 10% AS solution were shown to be unstable, since they visibly separated into phases about 1 h after preparation. However the emulsions formed with the other concentrations were stable for 4 h with the three sweetener:oil ratios used. Thus the emulsion with 5% AS and a sweetener:oil ratio of 1:1 was chosen for the production of microcapsules. Six formulations of microcapsules were produced, differing from one another with respect to the concentrations of the GE and GA solutions and the ratio of the core material (emulsion of AS in oil) in relation to the total mass of wall material. The formulations were denominated as follows: A: 2.5% GE & GA and 50% of core material; B: 2.5% GE & GA and 75% of core material; C: 2.5% GE & GA and 100% of core material; D: 5.0% GE & GA and 50% of core material; E: 5.0% GE & GA and 75% of core material; F: 5.0% GE & GA and 100% of core material.

Microcapsules were obtained with all the formulations tested, and as can be seen in Fig. 1A–C, were multinucleated (which confers characteristics of matrix and reservoir), with droplets of AS emulsion distributed at the centre of the microcapsules and not in the wall, which confers excellent controlled release characteristics to the capsules (Dong et al. 2011), which is one of the main objectives of encapsulating sweeteners. It can also be seen that as the amount of core material increased, so the microcapsules became less spherical, as also observed by Dong et al. (2011).

The electronic microscope images were similar for all the systems studied. Fig. 1D shows that the microcapsules had continuous walls showing no cracks or apparent porosity, which indicates the freezing and freeze-drying processes were adequate, since they did not damage the particles. Whole continuous walls are important for microcapsules, to assure greater protection and retention of the encapsulated material.

The microcapsules were connected to one another by solid bridges, an effect also observed by Prata, Zanin, Ré, and Grosso (2008) when encapsulating vetiver oil by complex coacervation

Table 1
Analytical determinations of the freeze-dried coacervated aspartame microcapsules.

Formulation	Hygroscopicity (g/100 g)	Moisture (%)	Solubility (%)	Particle size (μm)	Encapsulation yield (%)
A	13.43 ± 0.94a	5.86 ± 0.23^{ab}	9.46 ± 1.34^{c}	96.29 ± 0.06	45.15 ± 1.86^{d}
B	12.88 ± 1.42^{a}	7.42 ± 1.46^{a}	21.37 ± 0.95^{a}	99.76 ± 0.05	48.91 ± 2.22^{d}
C	12.29 ± 0.86^{a}	4.24 ± 0.11^{b}	18.95 ± 3.71^{ab}	102.38 ± 0.05	55.86 ± 1.35^{c}
D	10.73 ± 0.25^{a}	5.16 ± 0.42^{ab}	16.5 ± 3.19^{b}	95.40 ± 0.05	63.94 ± 0.23^{b}
E	12.23 ± 0.48^{a}	4.27 ± 0.21^{b}	13.81 ± 0.42^{b}	84.22 ± 0.11	71.70 ± 0.58^{a}
F	12.41 ± 2.63^{a}	3.99 ± 0.66^{b}	19.95 ± 1.93^{ab}	90.92 ± 0.05	64.11 ± 1.34^{b}
AS	10.86 ± 0.03^{a}	5.56 ± 0.01^{ab}	93.94 ± 0.57^{e}	–	–
GA	38.13 ± 1.73^{c}	9.17 ± 0.32^{c}	31.91 ± 1.71^{d}	–	–
GE	27.16 ± 0.06^{d}	11.99 ± 0.40^{d}	95.70 ± 0.62^{e}	–	v

^{a,b,c}Means with different small letters in the same column differ ($p < 0.05$).
A = 2.5% GE & GA and 50% of core material; B = 2.5% GE & GA and 75% core material; C = 2.5% GE & GA and 100% of core material; D = 5.0% GE & GA and 50% of core material; E = 5.0% GE & GA 5.0% and 75% of core material; F = 5.0% GE & GA and 100% of core material.

using GE and GA as the wall materials. These solid bridges can be attributed to agglomeration of the microcapsules caused by the process of freezing followed by freeze-drying.

3.2. Physicochemical characterisation of the coacervated microcapsules

Table 1 shows the values obtained for the mean size of the microcapsules, moisture content, and the solubility and hygroscopicity of the powders. The mean size of the particles varied from 84 to 102 μm, and although larger than the values observed by Mendanha et al. (2009) and Nori et al. (2011), are in agreement with other studies published in the literature, which cite variations between 1 and 500 μm for microcapsules produced by complex coacervation (Fávaro-Trindade et al., 2008). The concentration of the wall materials showed a slight influence on the size of the microcapsules, since the formulations A, B and C showed microcapsules with larger mean diameters than those from formulations D, E and F. Thus, the greater was the concentration of the wall material polymers in the formulation of the microcapsules, so the mean sizes of the microcapsules were slightly smaller. This influence was cited in studies which associated the sizes of the microcapsules with the production parameters, such as: proportion between the wall polymers, concentration of the polymers used, speed of agitation, rate of cooling and type of drying (Menger, Peresypkin, Caran, & Apkarian, 2000; Lamprecht, Schaè-Fer, & Lehr, 2001; Nakagawa, Iwamoto, Nakajima, Shono, & Satob, 2004, Silva, Fávaro-Trindade, Rocha, & Thomazini, 2012).

The moisture contents of the microcapsules were considered low, being within the range expected for freeze-dried products. Low moisture contents are desirable to guarantee the prevention of agglomeration, which reduces retention of the active principal and makes dispersion of the microcapsules difficult during the application in a food (Silva et al., 2012). In addition, low moisture contents reduce plasticiser action of water, which would reduce the glass transition temperature (Ferrari, Germer, Alvim, Vissotto, & Aguirre, 2012).

A comparison of the values for solubility obtained for the non-encapsulated AS with those of the microcapsules showed a reduction in this parameter of up to 10-fold. Low values for solubility are characteristic of microcapsules produced by complex coacervation and desirable in the encapsulation of sweeteners, since reduced solubility should contribute to retarding their release, providing a more gradual release and possibly prolonging the sensation of sweetness during chewing of the food containing the microcapsules.

The values obtained for hygroscopicity were in the range between 10.73 and 13.43 g water absorbed/100 g sample for the six formulations of microcapsules studied, with no significant differences between them. These values were considered to be low, making packaging and handling of the material easier. The values obtained in the present work were lower (by up to three times) than those obtained by Nori et al. (2011) for propolis microcapsules obtained by complex coacervation, using soy protein isolate and low methoxyl pectin as the wall materials, and where the variation in concentration of the materials used also caused no significant variations in this parameter. A comparison of these results could infer that the use of gelatine and gum Arabic as wall materials results in less hygroscopic materials.

The values obtained for EY varied between 45.2 and 71.7 (Table 1) and were a little lower than those observed by Jun-xia et al. (2011), when encapsulating orange oil using the complex coacervation technique with soy protein and gum Arabic as the wall materials. It can be seen that the values were significantly higher for the formulations produced with a 5% concentration of the wall material (D, E and F). An increase in concentration of the GE and GA solutions possibly produced more resistant walls, leading to greater EY values.

3.3. Fourier transformed infrared spectroscopy (FTIR)

Since the spectra obtained for all the formulations were similar, only the spectra obtained for the non-encapsulated AS and for the AS encapsulated with formulation (A) are presented in Fig. 2.

Gum Arabic is a polysaccharide with free carboxyl groups, conferring a negative charge on the molecule. In the spectra obtained for the encapsulated samples, the large peak formed at about 2900 cm^{-1} was attributed to the presence of carboxyl groups from the gum Arabic, indicating that not all the carboxyl groups were involved in the coacervation process. Nevertheless, peaks characteristic of amides can be visualised at about 1500–1640 cm^{-1}, confirming the formation of a coacervate, since during the complex coacervation process, the carboxyl groups of the polysaccharides interact with the amine groups of proteins forming a complex that contains amides.

An observation of the spectra of the non-encapsulated and encapsulated AS shows that peaks characteristic of the AS molecule, such as, for example, the moderately intense peak between 1700 and 1800 cm^{-1}, coming from the ester present in the composition of the AS, were maintained in the composition of the AS, indicating that the coacervation process did not degrade the AS.

3.4. Water sorption isotherms

The GAB, BET and Peleg mathematical models were evaluated for the fit of the data obtained, and the GAB model fitted them better. Table 2 shows that the GAB model was adequate to fit the experimental data, due to the elevated values obtained for r^2. An evaluation of the parameters fitted showed little variation between

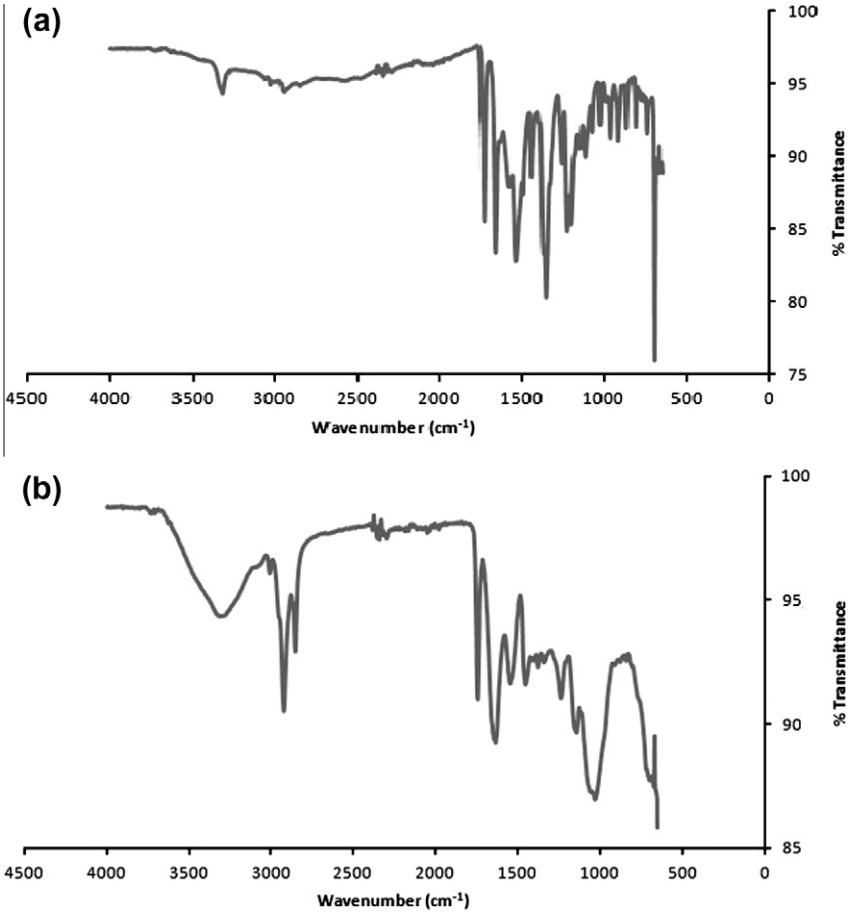

Fig. 2. Spectra obtained in the infrared region for (a) non-encapsulated aspartame and for (b) aspartame microencapsulated in formulation A (2.5% GE & GA and 50% of core material).

Table 2
Parameters for the fit of the GAB model to the isotherms of the aspartame microcapsules and the coefficients of determination (r^2).

Formulation	X_m	C	K	R^2
A	0.061	9.993	0.818	0.992
B	0.062	4.591	0.798	0.993
C	0.118	1.949	0.608	0.978
D	0.103	1.979	0.704	0.987
E	0.105	1.643	0.697	0.993
F	0.064	3.246	0.763	0.992

A = 2.5% GE & GA and 50% of core material; B = 2.5% GE & GA and 75% core material; C = 2.5% GE & GA and 100% of core material; D = 5.0% GE & GA and 50% of core material; E = 5.0% GE & GA 5.0% and 75% of core material; F = 5.0% GE & GA and 100% of core material.

the samples analysed. For BET and Peleg models were found r^2 values lower than 0.95.

No papers were found in the literature reporting on studies of the sorption isotherms of microcapsules produced by complex coacervation, which made it difficult to compare the results, which were obtained using spray dried powders. The parameter X_m (moisture content of the monolayer) corresponds to the amount of water strongly adsorbed to specific sites on the surface of the material, and is considered a critical value, above which the rates of the degradation reactions increase and the stability of the material is reduced. Low monolayer moisture contents were obtained, since the X_m varied from 6% to 11%. These values are similar to those obtained by Pérez-Alonso, Beristain, Lobato-Calleros, Rodriguez-Huezo, and Vernon-Carter (2006), who also obtained low monolayer moisture contents for pure gum Arabic (8.11–11.0%) at temperatures of 25, 35 and 40 °C.

With respect to the parameter K, which represents the correction factor for the properties of the multilayer molecules in relation to the product volume, low values were observed of between 0.6 and 0.8, within the values suggested by Lewicki (1997) of $0.24 < K < 1$. Values for K below 1.0 are characteristic of food products. Regarding the sorption constant C, which is due to interactions between the active sites of the product and the water molecules, it was observed that all the formulations showed values less than 200, being within the range commonly found in the literature (Alexandre, Figueirêdo, & Queiroz, 2007).

Fig. 3 shows the sorption isotherms for the AS microcapsules, showing an increase in equilibrium moisture content with increasing water activity. No studies were found in the literature concerning the isotherms of microcapsules produced by complex coacervation. However, the same behaviour was reported by Comunian et al. (2011), Catelam, Fávaro-Trindade, and Romero (2011) and Tonon, Brabet, Pallet, Brat, and Hubinger (2009), who constructed isotherms for chlorofilida, passion fruit and açai puree powders, all obtained by spray drying with maltodextrin as the wall material. Nevertheless the results obtained for moisture equilibrium in the present study were much lower than those reported by Comunian et al. (2011), Catelam et al. (2011) and Tonon et al. (2009), for all the water activity values, conferring greater stability, and ease of handling, storage and application, corroborating the previously discussed results obtained for hygroscopicity.

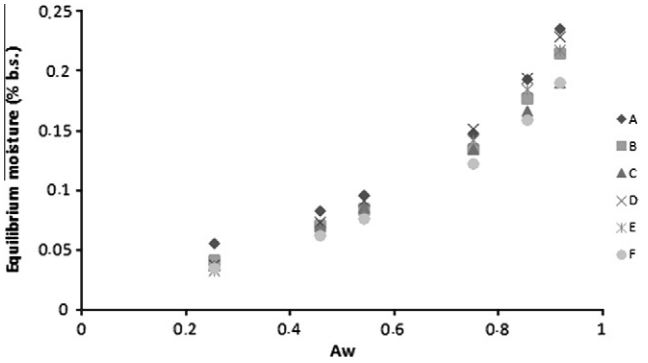

Fig. 3. Sorption isotherms of the microencapsulated aspartame samples fitted to the GAB mathematical model A = 2.5% GE & GA and 50% of core material; B = 2.5% GE & GA and 75% core material; C = 2.5% GE & GA and 100% of core material; D = 5.0% GE & GA and 50% of core material; E = 5.0% GE & GA 5,0% and 75% of core material; F = 5.0% GE & GA and 100% of core material.

3.5. Release of aspartame from the microcapsules into water at 36 and 80 °C

Fig. 4 shows the profile of the release of AS from the microcapsules into water at 36 and 80 °C for systems A, B and C, which all had the same concentration of wall material (2.5%), but differed with respect to the amount of core material.

The release profiles showed two phases, with the rate of release falling very quickly in the first phase, and then more slowly in the second phase. This behaviour was also observed by Dong et al. (2011), who analysed the profile of the release of mint oil from microcapsules into hot water.

For both temperatures it can be seen that the smaller the amount of core material, the greater the rate of release. This could be attributed to the particle size, since according to the results for particle size, the smaller the amount of core material, the greater the mean diameter, leading to a greater surface contact with the water, making diffusion of the core material easier and thus increasing the rate of release.

Analysing the two temperatures tested, it can be seen that an increase in temperature did not lead to an increase in the rate of release, showing that the microcapsules were relatively resistant to high temperatures (80 °C). This behaviour was expected for microcapsules produced by complex coacervation, and is important for their application in products where high processing temperatures are used, as for example chewing gum where sweeteners are usually used.

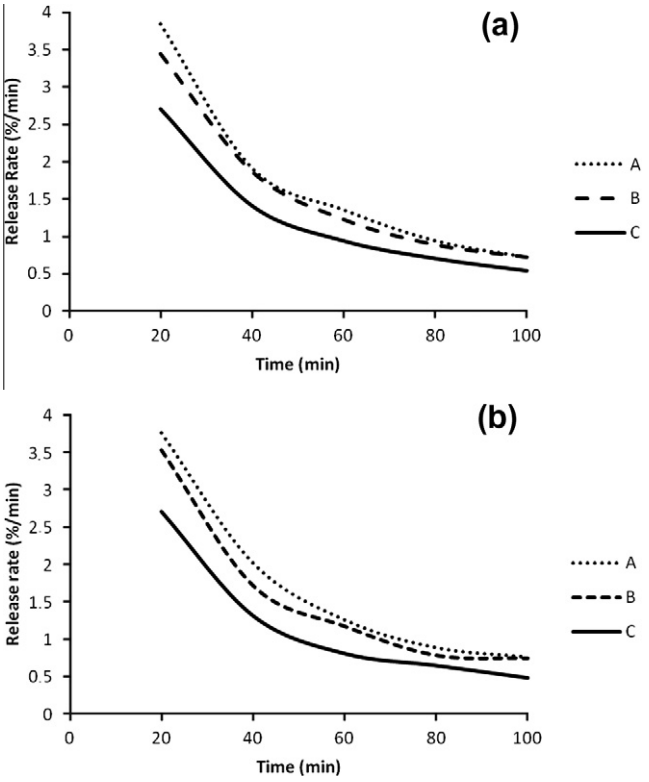

Fig. 4. Rate of release of aspartame from microcapsules with different amounts of core material (A = 50%, B = 75% and C = 199%) into water at 36 °C (a) and 80 °C (b).

4. Conclusions

Considering the proposed objectives and the results obtained, it can be concluded that the six formulations studied formed microcapsules with characteristics similar to those formed by complex coacervation, such as reduced solubility and heat resistance, indicating that the addition of a double emulsion to the process made it feasible to microencapsulate aspartame by this technique. In addition, the powder obtained was only slightly hygroscopic, making its application easier.

All microcapsules studied in this research showed the potential for application in food, especially the formulations D, E and F, which showed higher values of EY. Further studies should be carried out in order to evaluate the functionality of microcapsules in food products.

Acknowledgements

The authors thank FAPESP for the financial support (Process 2010/06146-9) and for the scholarship conceded to Fernanda Bozza (Process 2010/15215-4) and also to CNPq for the scholarship conceded to Glaucia Rocha-Selmi.

References

Alexandre, H. V., Figueirêdo, R. M. F., & Queiroz, A. J. M. (2007). Isotermas de Adsorção de Umidade de Pitanga em Pó. *Revista de Biologia e Ciências de Terra, Campina Grande, 7*(001), 11–20.

Butchko, H. H., Stargel, W. W., Comer, C. P., Mayhel, D. A., & Andress, S. E. (2001). Apartame. In L. O'B. Nabors (Ed.), *Alternative sweeteners* (3rd. ed. New York: Marcel Dekker Inc..

Cai, Y. Z., & Corke, H. (2000). Production and properties of spray-dried Amaranthus Betacyanin pigments. *Journal of Food Science, 65*(6), 1248–1252.

Cano-Chauca, M., Stringheta, P. C., Ramos, A. M., & Cal-Vidal, J. (2005). Effect of the carriers on the microstructure of mango powder obtained by spray drying and its functional characterization. *Innovative Food Science and Emerging Technologies, 5*(4), 420–428.

Catelam, K. T., Fávaro-Trindade, C. S., & Romero, J. T. (2011). Water adsorption isotherms and isosteric sorption heat of spray-dried and freeze-dried dehydrated passion fruit pulp with additives and skimmed milk. *Ciência e Agrotecnologia, 35*(6), 1196–1203.

Comunian, T. A., Monterrey-Quintero, E. S., Thomazini, M., Balieiro, J. C. C., Piccone, P., Pittia, P., et al. (2011). Assessment of production efficiency, physicochemical properties and storage stability of spray-dried chlorophyllide, a natural food colourant, using gum Arabic, maltodextrin and soy protein isolate-based carrier systems. *International Journal of Food Science & Technology, 46*, 1259–1265.

Dong, Z., Ma, Y., Hayar, K., Jia, C., Xia, S., & Zhang, X. (2011). Morphology and release profile of microcapsules encapsulating peppermint oil by complex coacervation. *Journal of Food Engineering, 104*(3), 455–460.

Fávaro-Trindade, C. S., Pinho, S. C., & Rocha, G. A. (2008). Microencapsulação de ingredientes alimentícios. *Brazilian Journal of Food Technology, 11*(2), 103–112.

Fávaro-Trindade, C. S., Santana, A. S., Monterrey-Quintero, E. S., Trindade, M. A., & Netto, F. M. (2010). The use of spray drying technology to reduce bitter taste of casein hydrolysate. *Food Hydrocolloids, 24*, 336–340.

Ferrari, C. C., Germer, S. P. M., Alvim, I. D., Vissotto, F. Z., & Aguirre, J. M. (2012). Influence of carrier agents on the physicochemical properties of blackberry powder produced by spray drying. *International Journal of Food Science & Technology, 47*(6), 1237–1245.

Gouin, S. (2004). Microencapsulation: industrial appraisal of existing technologies and trends. *Food Science and Technology, 15*, 330–347.

Jun-xia, X., Hai-yan, Y., & Jian, Y. (2011). Microencapsulation of sweet orange oil by complex coacervation with soybean protein isolate/gum Arabic. *Food Chemistry, 125*, 1267–1272.

Lamprecht, A., Schaè-Fer, U., & Lehr, C. M. (2001). Influences of process parameters on preparation of microparticle used as a carrier system for ω3 unsaturated fatty acid ethyl esters used in supplementary nutrition. *Journal of Microencapsulation, 18*(3), 347–357.

Lewicki, P. (1997). The applicability of the GAB model to food water sorption isotherms. *International Journal of Food Science and Technology, 32*, 553–557.

Mendanha, D. V., Ortiz, S. E. M., Fávaro-Trindade, C. S., Mauri, A., Monterrey-Quintero, E. S., & Thomazini, M. (2009). Microencapsulation of casein hydrolysate by complex coacervation with SPI/pectin. *Food Research International, 42*, 1099–1104.

Menger, F. M., Peresypkin, A. V., Caran, K. L., & Apkarian, R. P. A. (2000). Sponge morphology in an elementary coacervate. *Langmuir, 16*, 9113–9116.

Nabors, L. O. '. B. (2002). Sweet choices: sugar replacements for foods and beverages. *Food Technology, 56*(7), 28–34.

Nakagawa, K., Iwamoto, S., Nakajima, M., Shono, A., & Satob, K. (2004). Microchannel emulsification using gelatin and surfactant-free coacervate microencapsulation. *Journal of Colloid and Interface Science, 278*, 198–205.

Nori, M. P., Fávaro-Trindade, C. S., Alencar, S. M., Thomazini, M., Balieiro, J. C. C., & Castillo, C. J. C. (2011). Microencapsulation of propolis extract by complex coacervation. *LWT – Food Science and Technology, 44*, 429–435.

Pérez-Alonso, C., Beristain, C. I., Lobato-Calleros, C., Rodriguez-Huezo, M. E., & Vernon- Carter, E. J. (2006). Thermodynamic analysis of the sorption isotherms of pure and blended carbohydrate polymers. *Journal of Food Engineering, 77*(4), 753–760.

Prata, A. S., Zanin, M. H. A., Ré, M. I., & Grosso, C. R. F. (2008). Release properties of chemical and enzymatic crosslinked gelatin-gum Arabic microparticles containing a fluorescent probe plus vetiver essential oil. *Colloids and Surfaces B: Biointerfaces, 67*, 171–178.

Qv, X. Y., Zeng, Z. P., & Jiang, J. G. (2011). Preparation of a lutein microencapsulation by complex coacervation method and its physicochemical properties and stability. *Food Hydrocolloids, 25*, 1596–1603.

Ré, M. I. (1998). Microencapsulation by spray drying. *Drying Technology, 16*, 1195–1236.

Salminen, S., & Hallikainen, A. (2001). Sweeteners. In J. H. T. Iii (Ed.), *Food additives*. CRC Press.

Shahidi, F., & Han, X. Q. (1993). Encapsulation of food ingredients. *Critical Reviews in Food Science and Nutrition, 33*(6), 501–547.

Silva, D. F., Fávaro-Trindade, C. S., Rocha, G. A., & Thomazini, M. (2012). Microencapsulation of lycopene by gelatin–pectin complex coacervation. *Journal of Food Processing and Preservation, 36*, 185–190.

Tonon, R. V., Brabet, C., Pallet, D., Brat, P., & Hubinger, M. D. (2009). Physicochemical and morphological characterisation of açai (*Euterpe oleraceae* Mart.) powder produced with different carrier agents. *International Journal of Food Science and Technology, 44*, 1950–1958.

Wetzel, C. R., & Bellt, L. N. (1998). Chemical stability of encapsulated aspartame in cakes without added sugar. *Food Chemistry, 63*(1), 33–37.