Food Chemistry 171 (2015) 32–39

Contents lists available at ScienceDirect

Food Chemistry

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

Microencapsulation of xylitol by double emulsion followed by complex coacervation

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ARTICLE INFO

Article history:
Received 7 May 2014
Received in revised form 19 August 2014
Accepted 21 August 2014
Available online 30 August 2014

Chemical compounds studied in this article:
Xylitol (PubChem CID: 6912)
Phosphoric acid (PubChem CID: 1004)
Lithium chloride (PubChem CID: 433294)
Potassium sulphate (PubChem CID: 24507)
Potassium carbonate (PubChem CID: 11430)
Magnesium nitrate (PubChem CID: 25212)
Sodium chloride (PubChem CID: 5234)
Potassium chloride (PubChem CID: 4873)
Sodium hydroxide (PubChem CID: 14798)
Dibasic sodium phosphate (PubChem CID: 24203)

Keywords:
Release study
FTIR
Encapsulation efficiency
Sorption isotherms

ABSTRACT

The objective of this study was to produce and characterise xylitol microcapsules for use in foods, in order to prolong the sweetness and cooling effect provided by this ingredient. Complex coacervation was employed as the microencapsulation method. A preliminary double emulsion step was performed due to the hydrophilicity of xylitol. The microcapsules obtained were characterised in terms of particle size and morphology (optical, confocal and scanning electron microscopy), solubility, sorption isotherms, FTIR, encapsulation efficiency and release study. The microcapsules of xylitol showed desirable characteristics for use in foods, such as a particle size below 109 μm, low solubility and complete encapsulation of the core by the wall material. The encapsulation efficiency ranged from 31% to 71%, being higher in treatments with higher concentrations of polymers. Release of over 70% of the microencapsulated xylitol in artificial saliva occurred within 20 min.

1. Introduction

Xylitol is a polyol derived from xylose that has been widely used in the food industry due to its numerous benefits. Among them are its high sweetness, as a natural sweetener (Gliemmo, Calvino, Tamasib, Gerschensona, & Campos, 2008; Ikeuchi, Azuma, Kato, & Ooshima, 1999; Parajó, Dominguéz, & Dominguéz, 1998) and its high power of freshness due to its endothermic heat of dissolution (Mussatto & Roberto, 2002). Xylitol is considered to be anticariogenic (Gazzani, Daglia, & Papetti, 2012) and is therefore used as an ingredient in oral care products. In addition, studies have shown that xylitol can be used in the treatment of osteoporosis and respiratory disease (Mussatto & Roberto, 2002).

The microencapsulation process has been used for many years by the pharmaceutical industry, and is now an established technique. In recent years, microencapsulation has also been used in the food industry (Onwulata, 2012), for several purposes, such as to protect food ingredients from adverse factors, such as light, heat and humidity. Other advantages of microencapsulation include its ability to alter textures (transform liquids into powders), reduce volatility of essential oils, mask undesirable flavours and control the release of ingredients. In the case of xylitol, microencapsulation is desired to provide controlled release, in order to prolong the feeling of freshness and sweetness.

Various methods are employed in the production of microcapsules. In the food industry; the most common methods employed are spray drying, spray-chilling/cooling, fluidised beds, co-crystallisation, emulsification and complex coacervation. The process of complex coacervation is particularly suitable when the aim is to control the release of the agent, because this process produces a true capsule, completely protecting the active material within the wall material.

Complex coacervation involves the mixture of two polymers at a pH at which one polymer is negatively charged and the other is

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http://dx.doi.org/10.1016/j.foodchem.2014.08.093
0308-8146/

positively charged, leading to phase separation and the formation of insoluble particles in water (Lakkis, 2007).

One of the limitations of complex coacervation is the difficulty of encapsulating hydrophilic compounds, as this technique is more suitable for hydrophobic compounds as the filling material. To encapsulate hydrophilic compounds, it is necessary to make adjustments to the method, e.g., by including a double emulsion process (Mendanha et al., 2009).

While patents have described the microencapsulation of xylitol for use in foods (Wolf et al., 2008), no published studies on this topic are available. There is a study on xylitol encapsulation by interfacial polymerisation, but is not appropriate in food (Salaün, Bedek, Devaux, Dupont, & Gengembre, 2011). Therefore, the aim of this study was to produce microcapsules of xylitol, in order to control the release of this ingredient, thus prolonging its sweetness and freshness when applied in foods, such as in chewing gum.

2. Materials and methods

2.1. Materials

Xylitol was supplied by Danisco (Brazil), pork gelatin type B was provided by Gelita (Mococa, Brazil), arabic gum was provided by Nexira (São Paulo, Brazil), the corn oil was of the Liza brand (Brazil), the phosphoric acid was of the Synth brand (São Paulo, Brazil) and polyglycerol polyricinoleate (PGPR 90) was supplied by Danisco (Denmark).

2.2. Methods

2.2.1. Emulsions

For the simple emulsion (W/O) an aqueous solution of xylitol (1 g of xylitol/0.3 g of water at 80 °C) was added to corn oil (2 g/1 g aqueous xylitol solution) in the presence of PGPR (0.008 g/1 g total solution) and homogenised in an Ultraturrax homogenizer (T25-D model; IKA® Werke GmbH & Co., Staufen, Germany) for 4 min at 12,000 rpm. To prepare the double emulsion (W/O/W), the simple emulsion was slowly added to 100 mL of an aqueous gelatin solution (2.5; 5.0 and 7.5 g/100 mL of water) at 50 °C and homogenised for 3 min at 10,000 rpm. The parameters were defined in preliminary tests based on the study conducted by Comunian et al. (2013).

2.2.2. Complex coacervation

A gum Arabic aqueous solution (2.5; 5.0 and 7.5 g/100 mL of water) at 50 °C was slowly added to the double emulsion under constant magnetic stirring. Then, the pH was adjusted to 4.0 using phosphoric acid. Slow cooling was performed in an ice bath under stirring until the sample reached 10 °C, and then the coacervate material was stored for 24 h at 7 °C to promote decantation, facilitating the removal of the aqueous phase. To increase the sample stability and facilitate handling, the coacervated microcapsules were freeze-dried (Terroni, São Carlos, Brazil). The operation conditions were as follows: process time of 24 h, pressure of 1–0.1 kPa, condenser temperature of −20 °C and final temperature of 30 °C (Comunian et al., 2013). Different polymer concentrations and cores were analysed, totalling nine treatments. The formulations evaluated are described in Table 1.

2.2.3. Characterisation of xylitol microcapsules

2.2.3.1. Particle size. A laser diffraction particle analyser was used to determine the particle size (SALD-201V; Shimadzu, Kyoto, Japan). The wet microcapsules (before freeze-drying) were dispersed in distilled water and placed in the glass bottle provided with the equipment for analysis. Three measurements were performed and the operation was repeated three consecutive times for each sample. The average particle size was represented as the volume-weighted mean diameter.

2.2.3.2. Optical microscopy. The morphology of the emulsions and microcapsules were studied by an optical microscope (Bel Photonics BIO3; BEL Engineering srl, Monza, Italy) equipped with a camera, using the V.62 BEL View software for image analysis. The wet microcapsules (before freeze-drying) were dispersed on a glass slide, which was fixed in the microscope for observation.

2.2.3.3. Confocal microscopy. The samples were examined in the National Institute of Science and Technology on Photonics Applied to Cell Biology (INFABIC) at the State University of Campinas, using a Zeiss LSM 780-NLO confocal microscope and Axio Observer Z.1 microscope (Carl Zeiss AG; Jena, Germany) using a 40× objective. Images were collected using the 561 nm laser line for excitation and 569–691 nm emission filters for Nile red fluorophores, with pinholes set to 1 Airy unit for each channel, in a 1024 × 1024 image format, with a 1.7× optical zoom.

2.2.3.4. Scanning electron microscopy (SEM). The freeze-dried microcapsules were adhered to double-sided carbon tape, set in aluminium stubs. The images were captured on a scanning electron microscope (Hitachi Tabletop Microscope, Japan) with an accelerating voltage of 5 kV and a current of 1750 mA.

2.2.3.5. Solubility. A sample of 0.50 g was added to an Erlenmeyer flask containing 50 mL of distilled water and homogenised on an orbital shaker table at 100 rpm for 30 min at room temperature. Then, the solution was centrifuged at 3500 rpm for 5 min. An aliquot of 25 mL of the supernatant was transferred to a porcelain capsule with a known mass and maintained at 105 °C until a

Table 1
Microcapsule formulations, particle sizes, solubilities and encapsulation efficiencies (*EE*).

Formulation	Polymers (*w*/*v*)*	Core (*w*/*w*)**	Particle size (μm)	Solubility (%)	*EE* (%)
T1	2.5	50	104.69 ± 10.78^{ab}	5.31 ± 0.82^{f}	44.69 ± 12.12^{bcd}
T2		100	109.31 ± 6.57^{a}	6.25 ± 1.39^{ef}	32.84 ± 5.84^{cd}
T3		150	103.97 ± 3.98^{ab}	7.96 ± 2.04^{de}	31.42 ± 5.61^{d}
T4	5.0	50	78.45 ± 7.78^{d}	8.65 ± 1.25^{cd}	44.27 ± 10.67^{bcd}
T5		100	90.21 ± 8.54^{c}	8.96 ± 0.92^{cd}	47.57 ± 14.64^{b}
T6		150	95.07 ± 4.05^{bc}	10.28 ± 1.30^{bc}	46.11 ± 9.18^{bc}
T7	7.5	50	90.32 ± 7.71^{c}	11.37 ± 1.65^{b}	71.93 ± 9.19^{a}
T8		100	92.36 ± 5.05^{c}	11.92 ± 0.41^{ab}	62.94 ± 8.03^{a}
T9		150	93.29 ± 5.84^{c}	13.90 ± 2.09^{a}	62.13 ± 6.65^{a}

Means ± standard deviation values in the same column followed by the same superscripts are not significantly different ($p < 0.05$). $n = 9$.
* The ratio of gelatin:arabic gum was fixed at 1:1.
** In relation to the amount of polymer.