Journal of Food Engineering 146 (2015) 234–242

Contents lists available at ScienceDirect

Journal of Food Engineering

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

Compressed tablets based on mineral-functionalized starch and co-crystallized sucrose with natural antioxidants

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ARTICLE INFO

Article history:
Received 17 June 2014
Received in revised form 1 September 2014
Accepted 3 September 2014
Available online 19 September 2014

Keywords:
Compressed tablets
Starch
Zinc
Yerba mate
Co-crystallization
Delivery system

ABSTRACT

Compressed tablets based on starch–zinc carriers and co-crystallized sucrose with yerba mate antioxidants were developed. Firstly, the zinc-binding capacity of corn starch was evaluated for being used as a mineral source for the formulations. While, the yerba mate extract was entrapped within a sucrose matrix by co-crystallization. Then, tablets were obtained by compression using co-crystallized products alone and their blends with starch-carriers (85:15, 80:20 and 70:30). All these formulations led to higher tablet hardness than equivalent blends containing raw sucrose. However, high dosages of starch-carriers provoked defects in the tablets (e.g. "capping"). The weight ratio of 80:20 allowed obtaining tablets with optimal hardness values (45–55 kPa), containing zinc (5.4 mg/g tablet) and yerba mate polyphenols (1.3 mg/g tablet). These tablets showed low disintegration times (<10 min) and a fast-release in aqueous medium constituting a useful way for the oral delivery of active compounds with health benefits.

1. Introduction

The design of products that promote health benefits beyond to providing the nutritional and energetic requirements constitutes an actual challenge for the food industry. This trend has led to increase the use of functional ingredients such as antioxidants, minerals, peptides, vitamins, fatty acids, probiotic, among others. However, the incorporation of some compounds in their original state is not always possible due to their unpleasant flavor, low stability or bioavailability; therefore, a previous process to face up these disadvantages is often necessary.

Yerba mate (*Ilex paraguariensis*) is a source of bioactive ingredients (mainly polyphenols, xanthines, flavonoids, saponins, amino acids, minerals and vitamins) with potential application in food and pharmaceutical industries. These compounds may be extracted and concentrated to be used as natural additives (colorant, antioxidant, antimicrobial and/or stimulant), as functional ingredients (hepatoprotective, diuretic, hypocholesterolemic, antirheumatic and antithrombotic) and/or as nutritional supplements in tablets or capsules (Marques and Farah, 2009; Bracesco et al., 2011; Racanicci et al., 2011).

Zinc (Zn) is an essential micronutrient for human growth, development and function of the immune system (Salgueiro et al., 2000; Tapiero and Tew, 2003). Besides, Zn has antioxidant properties which could avoid the illness appearance from oxidative stress (Powell, 2000; Zago and Oteiza, 2001; Goel et al., 2005). Nevertheless, Zn deficiency is one of the ten biggest factors contributing to the burden of disease in developing countries with high mortality (WHO, 2002; Shrimpton et al., 2005). Zinc sulfate is commonly used as a Zn source for supplementation due to its low cost and bioavailability (Salgueiro et al., 2000). However, several authors have reported that this compound modifies the product sensorial characteristics rendering flavor unacceptable and it can also generate side-effects such as nausea and vomiting (Salgueiro et al., 2002; Solomons et al., 2011).

The advantage of using a combination of antioxidants and minerals was reported by several authors, since these compounds could act individually, cooperatively and synergistically (Hercberg et al., 2004; Carocho and Ferreira, 2013). Therefore, the development of systems for the simultaneous carrying from zinc and yerba mate antioxidants is a useful strategy to supply the nutritional requirements. However, both active compounds are chemically reactive and their functionality could be affected by interactions with other components of the vehicle matrix. In this sense, encapsulation is a useful way to protect sensitive materials

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http://dx.doi.org/10.1016/j.jfoodeng.2014.09.019
0260-8774/

against adverse conditions and to mask unpleasant flavors (Day et al., 2009). In a previous work, the compartmentalization in a alginate–starch matrix proved to be an effective tool for the simultaneous transport of zinc and yerba mate antioxidants, preventing a possible interaction between them (López-Córdoba et al., 2014b).

Co-crystallization represents a viable means of enhancing the physical properties of active compounds such as solubility, dispersibility, wettability, anticaking, antidusting, antiseparation, homogeneity, flowability and stability (Bhandari and Hartel, 2002). In this process, the crystalline structure of sucrose is modified from perfect to irregular agglomerated crystals, to provide a porous matrix in which a second active ingredient can be incorporated. Because of their agglomerated structure, all co-crystallized sugar products offer direct tableting characteristics which provide significant advantages in the candy and pharmaceutical industries (Awad and Chen, 1993; Chen et al., 1988).

Nowadays, the compressed tablets are the most popular dosage form used; they are composed of active ingredients and excipients (e.g. binders, disintegrants, lubricants and diluents). Tablets have many advantages over other dosage forms including competitive unit production costs, manufacturer simplicity, good portability and easy administration. Moreover, they offer better stability to heat and moisture compared to liquid and semi-solid formulations (Jivraj et al., 2000).

Starch is a natural, renewable, biodegradable polysaccharide composed primarily of branched and linear chains of glucose molecules, named as amylopectin and amylose, respectively (Fennema and Tannenbaum, 1996). Starch is considered a good diluent, disintegrant, tablet binder and thickening agent used in pharmaceutical dosage formulations (Jivraj et al., 2000).

The functionalization of starch with bioactive compounds constitutes a new approach for the formulation of healthy products. Several authors have reported the ability of native and modified starches to embed guest molecules such as drugs, transition metals, flavors, vitamins, among others (Zhao et al., 1996; Lii et al., 2002; Staroszczyk and Janas, 2010; Ades et al., 2012; Luo et al., 2013; Janaswamy, 2014). In the present work, starch-carriers and co-crystallized products were combined to obtain compressed tablets with zinc and yerba mate antioxidants. Also, the Zn-binding capacity of native corn starch and its possible application as a release agent were tested. The formulations of the compressed tablets were optimized and the products were characterized, as well. To our knowledge, it is the first time that a delivery system is developed using this combination of materials and techniques.

2. Materials and methods

2.1. Active compounds

Dried and minced yerba mate leaves were used as a source of natural antioxidants. An aqueous extract was prepared according to the previously optimized methodology by Deladino et al. (2008). Briefly, a blend of 10 g of commercial yerba mate (Las Marías, Corrientes, Argentina) and 100 mL of distillated water was placed in a thermostatic bath (Viking, Argentina) at 100 °C for 40 min. After this time, samples were filtered and cooled. Zinc sulfate·$7H_2O$ (Parafarm, Argentina) was used as a source of zinc (4.4 mg of zinc sulfate provided 1 mg of elemental zinc).

Both ingredients are listed as GRAS (generally regarded as safe) by Food and Drug Administration (FDA).

2.2. Co-crystallized sucrose with yerba mate extract

The co-crystallized products with yerba mate extract were prepared as described by López-Córdoba et al. (2014a). Briefly, a blend of raw sucrose (50 g) (Ledesma, Argentina) and yerba mate aqueous extract (10 mL) was heated to 132 °C on a hot plate and stirred with a vertical agitator (IKA Labortechnik, Staufen, Germany). When a slight turbidity was detected in the syrup, indicating the beginning of the crystallization process, the mix was removed from the heat, maintaining the agitation. The co-crystallized products were dried in a convection oven (SanJor, Argentina) at 40 °C for 15 h and then they were ground and sieved through a 500 μm mesh. Blends of raw sucrose (50 g) and distillated water (10 mL) were crystallized as described above for control purposes. These samples will be referred as "re-crystallized sucrose".

The content of yerba mate polyphenols loaded in the co-crystallized products was determined by Folin–Ciocalteu method as reported in a previous work (López-Córdoba et al., 2014a). The co-crystallization yield (%) was calculated as the ratio between the amount of yerba mate polyphenols loaded per gram of co-crystallized product and the mass of yerba mate polyphenols employed in the formulation per gram of raw sucrose.

2.3. Starch–zinc carriers

The starch–zinc carriers were prepared as described in a previous work (López-Córdoba et al., 2014b). Briefly, blends of native corn starch (1 g) (Unilever, Spain), deionized water (10 mL) and zinc sulfate (1.43 g/g of starch) were prepared under continuous stirring (180 rpm, 25 °C for 15 h). Then, the samples were centrifuged (at 5000g for 15 min), the supernatant was removed and the solids were dried and milled. The Zn adsorption yield (%) on starch was calculated as the ratio between Zn mass per gram of starch-carrier and the Zn mass employed in the formulation per each gram of native corn starch. In all cases, the Zn concentration was quantified by atomic absorption spectroscopy using the flame method with a wavelength of 213.9 nm. The analyses were carried out in a Varian spectrometer model EspectrAA 300-plus (Cambridge, United Kingdom). Previously, the samples were digested with concentrated nitric acid.

2.4. Characterization experiments of the powder materials

Starch–zinc carriers were analyzed by confocal laser scanning microscopy (CLSM), intact starch granules and cross-sections obtained by cryo-fracture were tested. Fluorescein isothiocyanate (FITC) (0.3 mg/mL) was used for labeling. Starch (5 mg/mL) were suspended in Milli-Q water, then 1000 μL of starch suspensions were stained by the addition of 40 μL of FITC. The mixture was agitated in a vortex and let to rest for 1 h in closed eppendorfs in darkness at room temperature before analysis. A LEICA TCS SP5 (Mannheim, Germany) inverted microscope equipped with Ar and HeNe laser was used. The excitation wavelength was 488 nm and the emission wavelength 518 nm. Images were acquired using a HCX PL APO CS 63.0 × 1.40/UV/oil immersion objective and with 1024 × 1024 pixel resolution in a constant z-position. Software Leica Application Suite Advanced Fluorescence (LAS AF), version 2.2.1. build 4842 was employed in the image analysis. Micrographs of co-crystallized products and re-crystallized sucrose were acquired by scanning electron microscopy (SEM) using an FEI, Quanta 200 equipment (The Netherlands). Samples were attached to stubs using a two-sided adhesive tape, then coated with a layer of gold (40–50 nm) and examined using an acceleration voltage of 20 kV.

Moisture content (%) was measured gravimetrically by drying the grounded samples in a vacuum oven at 70 °C, until constant weight (AOAC, 1998).

X-ray diffraction (XRD) analysis was performed in an X'Pert PRO (The Netherlands) equipment at 40 kV with radiation of wavelength of 40 mÅ. Samples were scanned with 2θ between 5° and