Encapsulation of Antioxidants by Spray-Drying

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This study provides an encapsulation process of at least one antioxidant in a carbohydrate matrix, by homogenization and spray-drying. The method involves homogenization of the antioxidant with the carbohydrate matrix in water, at controlled speed and temperature, followed by spray-drying under well established conditions. The process parameters used in the spray-drying are all selected and/or manipulated to produce an encapsulated antioxidant product of very fine particle size of 0.08±0.02 microns analysed by Scanning Electron Microscopy (SEM) and Laser Scattering. Such encapsulated antioxidant products are very stable, protected from oxidation and are useful for being incorporated into beverages, such as near-waters, supplements products, pharmaceutical and cosmetic products.

Introduction

Microencapsulation/Nanoencapsulation is defined as a technology of packaging solids, liquids or gaseous materials in miniature, sealed capsules that can release their contents at controlled rates under specific conditions (Desai and Park, 2005). These capsules protect the encapsulated product from the light and oxygen and consequently prevent its degradation. Antioxidants can be added to systems where they might degrade, become hazardous by oxidation reactions and change the original physical properties. Nanoencapsulation is one technology that can be used to overcome these problems and also permit the dilution of small amounts of the active compound in an uniform dispersion. Nanocapsules with a polymeric shell matrix, based on polysaccharides, were developed by spray-drying technology to encapsulate antioxidants from tea. The efficiency of the nanoencapsulation process was tested by different techniques and the results obtained show that the objectives were achieved: quantification of the encapsulated antioxidant relatively to the shell material; protection of the antioxidant from the environment; stability of the encapsulated antioxidant for a long period of time; release of the antioxidant from the shell under specific conditions. This process is particularly suitable for encapsulation of natural antioxidants that will be used in food, pharmaceutical and cosmetic industries. In these areas of application it is a challenge to use natural antioxidants in contact with air or water maintaining the antioxidant activity. These compounds including flavonoids are effective antioxidants because of their free radical scavenging properties and because they are chelators of metal ions (Kandaswami and Middleton, 1994). They may protect tissues against free oxygen radicals and lipid peroxidation. Their biological activities include antithrombotic, vasodilatory and anticarcinogenic effects, as well as anti-inflammatory, antiallergic, antiulcer, antibacterial and antiviral properties (Middleton et al., 2000).

Materials and Methods

This work provides a composition comprising at least one antioxidant encapsulated in a carbohydrate matrix, by a process of homogenization and spray-drying. The matrix composition is based on maltodextrins (percentages and composition are patent pending). The method of this work involves the homogenization of at least one antioxidant with the carbohydrate matrix in water, at controlled speed and temperature, followed by spray-drying the mixture. The process parameters used in spray-drying are all selected and/or manipulated to produce an encapsulated antioxidant product of very fine particle size, suitable for quick and complete dissolution, and also having an uniform particle size which provides for its uniform distribution in the preparation.

Materials

High molecular weight film forming carbohydrate was purchased from Sigma-Aldrich Co. (USA). Maltodextrins were kindly provided by Grain Processing Corporation (USA). Catechins were provided by Taiyo Kaguku (Japan). All chemicals were used without further purification. The water was deionized using an EasyPure RF (Barnstead GmbH) purification system.

Methods

Preparation of catechins encapsulated nanoparticles by spray-drying: The high molecular weight film forming carbohydrate was dissolved in water in a reaction vessel, adding the maltodextrin with stirring for an appropriate amount of time to form an aqueous solution with the temperature being maintained at around 50 to 60 ºC. The antioxidant was then added to the solution, the solution being preferably maintained at from 10 to 40 ºC. The solution was homogenized at a constant speed from 9000 to 10000 rpm with a dispersing device IKA DI25 Basic. The solution was then spray-dried in a pilot spray-dryer (designed by Niro A/S) wherein the inlet temperature was typically between 150 - 190 ºC.

Zeta Potential: The zeta potential was determined using a Malvern Zetasizer 5000 (Malvern Ltd, UK). Each sample was measured at a concentration of 0.5 wt.% in water at pH 5.

Laser Scattering: The size distribution by volume and by number was assayed by dispersing the antioxidant encapsulated nanocapsules in ethanol at 4 wt.% and measured on a Coulter LS 230 laser particle sizer. The laser sizer has PIDS (polarization intensity differential scattering) capability for measuring submicron particles.

Scanning Electron Microscopy (SEM): The surface morphology of the spray-dried carbohydrate nanocapsules loaded with an active compound was examined by means of a JEOL SEM 6301F microscope at the facilities of the Centre for Materials Engineering (CEMUP). The powders were previously fixed on a brass stub using double-sided adhesive tape and then were made electrically conductive by coating, in a vacuum, with a thin layer of platinum. Photographs were taken at an excitation voltage of 7 kV.

Confocal Laser Scanning Microscopy (CLSM): Fluorometric detection of encapsulated catechins was performed using a CLSM TCS Leica at the Institute for Molecular and Cell Biology. The maximum excitation wavelength of the fluorophore generated in the reaction with catechin and boric acid was 458 nm and the maximum emission wavelength was 550 nm.

UV-Vis Spectroscopy: The catechin content at 90ºC and at acidic pH values (pH 3 and 5) was measured using a T80 UV/VIS Spectrometer (PG Instruments Ltd).

Results and Discussion

The present study demonstrates the preparation of nanoparticles comprising catechins, encapsulated in a carbohydrate matrix. The carbohydrates, such as starches and maltodextrins have properties such as low viscosities at high solids contents and good solubility that are desirable in an encapsulating agent. In addition their diversity, low cost and widespread use in foods and drinks makes them the preferred choice for encapsulation (Desai and Park, 2005). The oral bioavailability of tea catechins is less than 2-5% (Dvorakova et al., 1999, Catterall et al., 2003) and their systemic clearance is high (Dvorakova et al., 1999, Cai et al., 2002). The encapsulation of tea catechins improves their bioavailability, prevents them from oxidation and increases their absorbency by controlled release. The encapsulated catechins can be used in various applications, including, for example, in drinks and foods as dietary supplements or in health care products. The encapsulation process comprises the steps of: forming an aqueous carbohydrate solution, with stirring for an appropriate amount of time and at sufficient temperature to form a homogeneous solution; incorporating at least one active compound defined above into the previous solution; spray-drying the aqueous solution at different temperatures to obtain a stable product encapsulated within the active compound. SEM pictures of spray-dried active compound-loaded carbohydrate nanoparticles are presented in Figure 1. It can be seen that the particles are spherical and have a smooth surface.

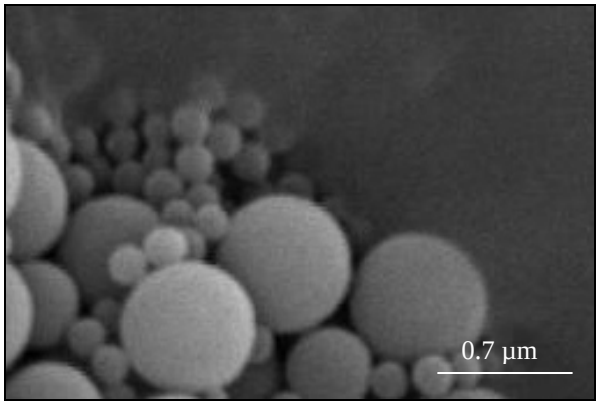

Figure 1. SEM picture of the catechin encapsulated nanocapsules.

The particle size distribution was determined by Laser Scattering and is depicted in Figure 2. The mean diameter was quantified to be 0.08±0.02 microns. The nanocapsules were negatively charged with a mean zeta potential of -22 mV. These results reveal that nanocapsules are stabilized ionically.

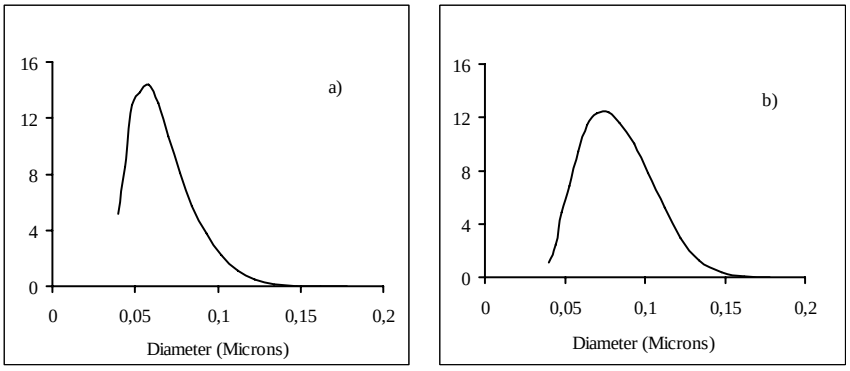

Figure 2. Nanocapsules size distribution by volume (a) and by number (b).

The confocal micrograph of nanocapsules in fluorescence mode shows that the catechins are concentrated in the core of the particles (Figure 3).

Figure 3. Confocal slicing of encapsulated catechins. The scale bar is 1 µm.

The release of catechins from the nanocapsules at pH 5 was observed by UV-VIS spectroscopy after heating the solution at temperature above 80ºC (Figure 4). It can be observed that the content of catechins detected by UV-VIS spectroscopy increases after heating the solution. This indicates that the encapsulation of catechins was successful and that they are released at temperatures above 80ºC. The stability of the nanocapsules is not affected by decreasing the pH. It can be seen from Figure 4 that the catechin content is the same at pH 3 and 5. This might be particular important when considering sterilization processes of drinks and food products. Moreover this result suggests that the nanocapsules might be stable at stomach pH. At pH 3, similar to pH 5, the antioxidant concentration increases at temperatures above 80ºC.

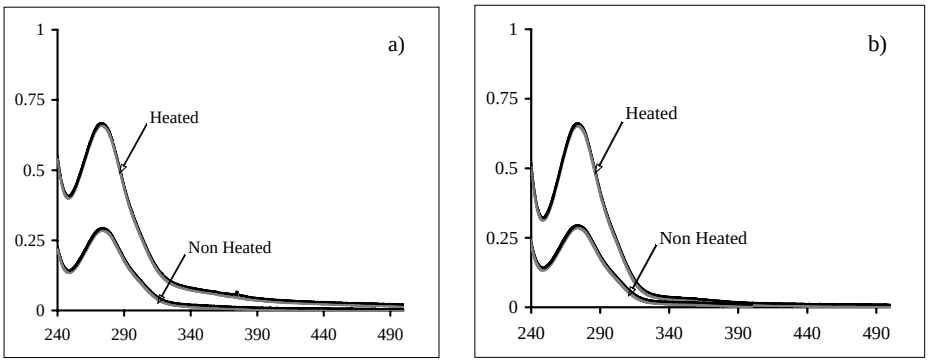

Figure 4. UV spectra of nanocapsule suspension before and after heating (90ºC) at: a) pH 5 and b) pH 3.

Conclusions

Nanoparticles containing catechins encapsulated in a carbohydrate matrix were produced by spray-drying. The particles are spherical with a smooth surface and a mean diameter of 0.08±0.02 microns. The zeta potential of the nanocapsules is highly negative, which contributes for their stabilization. The release of the core material is triggered by temperature (above 80 ºC), but not by acidic pH.

The encapsulation of catechins prevents them from oxidation and may improve their bioavailability. The nanoparticles described in this paper can be used as supplements and incorporated into functional food and beverages.

The results of this work are the basis of the encapsulation of other antioxidants, such as vitamins and carotenoids, using a similar carbohydrate matrix by spray-drying.

Acknowledgements

CIRES Company is gratefully acknowledged for technical support. The authors acknowledge Luso-American Foundation (FLAD) and UNICER Company for their motivation. Sandra Rocha thanks FCT for a fellowship (BDP/25641/2005). This work was partially supported by Agência de Inovação (ADI) under project ACTIVECAP and European Union under the project AGRIVAL (Interreg IIIB).

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