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Characterization of co-crystallized sucrose entrapped with cardamom oleoresin

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ABSTRACT

Cardamom oleoresin emulsified with gum acacia was encapsulated by co-crystallization in a supersaturated sucrose solution to prepare flavoured sucrose cubes. The co-crystals with and without cardamom oleoresin were characterized at 25°C and 33%, 63% and 93% relative humidity for hygroscopicity and crystallinity *vis-à-vis* pure crystal sucrose. Co-crystallized sucrose cubes showed lower hygroscopicity at 93% relative humidity, a longer dissolution time, and a decreased crystallinity as compared to crystal sucrose. The active components of cardamom oleoresin such as 1,8-cineole (30.23%) and α-terpinyl acetate (46.42%) in cardamom oleoresin was quantified by gas chromatography. The encapsulation efficiency of 1,8-cineole and α-terpinyl acetate in lab-made co-crystallized sucrose cubes was approximately 35.23% and 67.18%, respectively. This approach could contribute to value addition of cardamom oleoresin for flavoured tea and also have potential applications in traditional Indian sweets.

1. Introduction

Encapsulation of food flavours and various bioactive compounds is a well developed and interesting area in the field of food technology. Commercial production of these encapsulated actives is accomplished by a variety of methods like spray-drying, extrusion, co-acervation and adsorption.

Encapsulation by co-crystallization in sucrose matrix is a relatively new and simple method which offers an economical and flexible alternative for handling and preserving various active components used in the food industry (Jackson and Lee, 1991). In this process, the active ingredient is incorporated in supersaturated sucrose syrup to attain simultaneous crystallization of both components as well as entrapment of active ingredient in the sucrose matrix, respectively (Hartel, 1993). On co-crystallization, the crystalline structure of sucrose is modified from a perfect to an irregular agglomerated crystal to provide a porous matrix where the active ingredient can be incorporated (Chen et al., 1988). Careful incorporation of the active ingredient inhibits premature sucrose crystallization which permits the process to proceed at manageable and reproducible rates, and also improves the functionality of the co-crystallized product (Bhandari et al., 1998). Moreover, crystallization of the active ingredient may also occur during storage depending upon temperature and moisture conditions. In general, co-crystallization improves solubility, wettability, homogeneity, dispersibility, hydration, anti-caking and stability of the encapsulated materials (Beristain et al., 1996; Vazquez and Beristain, 1998). Co-crystallization to retain flavour compounds of honey, Jamaica granules and orange peel oil are reported by Bhandari et al. (1998), Beristain et al. (1994) and Beristain et al. (1996), respectively.

The active compound studied in the present work was cardamom oleoresin. Cardamom is known as the 'Queen of Spices' and is considered as the most expensive spice after vanilla and saffron. It finds its place in every kitchen as well as in pharmaceutical/ayurvedic medicines and cosmetics. India is the second largest producer of cardamom after Guatemala with an average production of 9000–12,000 tons followed by world production of 30,000–32,000 tons in the year 2010–11. At present, India is the second largest consumer of 11,000 MT cardamom in the world after Saudi Arabia with global consumption of 15,000–24,000 tons (http://www.karvycomtrade.com/disclaimer.asp).

Oleoresins represent a concentrated form and possess high replacement values which are specially suited for high temperature applications. Unlike powdered & liquid spices it offers uniformity and proper mixing of the corresponding components when incorporated in different food products. It exhibits pungency, cool, spicy, burning flavour and functionality in food processing applications (Farrell, 1985). Oleoresins contain natural antioxidants of the corresponding spices which make them more stable (Eiserle and Rogers, 1972). Moreover, the resin part in the oleoresins acts as natural fixatives to more volatile components. Cardamom oleoresins contain essential volatile oils (60%) containing isoprenoids and benezenoids, and non-volatiles (40%) containing fixed oils, alkaloids, carotenoids and anthocyanins (Boelens and Boelens, 2000). Two major active component of cardamom oleoresin are 1,8-cineole and α-terpinyl acetate. Terpenoids, more specifically

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α-terpinyl acetate in cardamom oil could undergo hydrolysis, rearrangement, polymerization, and oxidative reactions and thereby affect flavour due to its vulnerability to acid, light, oxygen or heat. These changes increase the content of *p*-cymene which is also a terpene but with petroleum-like aroma (Brennand and Heinz, 1970). These changes can be arrested by proper encapsulation of cardamom oleoresin.

The present work was undertaken to characterize the co-crystallized cardamom flavoured sucrose cubes containing emulsified cardamom oleoresin with gum acacia in a supersaturated sucrose solution. Since India is the largest producer of sucrose and second largest producer of cardamom throughout the world, it is worthwhile to develop value-added products from these indigenous bioresources that would not only have domestic but also international market. Such cardamom flavoured sucrose cubes could be used to make flavoured teas and also used in traditional Indian sweets.

2. Materials and methods

2.1. Materials

Food grade crystal sucrose (Sahakari Bhandar, Reliance Retail, Mumbai) and finely ground sucrose (M.B. Sucroses, Malegaon, Maharashtra, India) was used as the encapsulating raw material. Food grade gum acacia powder (Sigma Aldrich Ltd., Mumbai, India) was used as an emulsifying agent. Cardamom oleoresin was obtained as a gift sample from Plant Lipids Ltd., Cochin, India. Standard 1,8-cineole and α-terpinyl acetate was gifted from Simrise Private Limited, Mumbai and internal standard pentadecane was purchased from Merck Chemicals, Mumbai, India. Gas chromatography grade hexane was procured from Sd-fine Chemicals, Mumbai, India and analytical grade ethanol from Changshu Yangyuan Chemicals, China. All chemicals used were of AR grade.

2.2. Sample preparation

This was adapted from the paper by Bhandari and Hartel (2002). Experiments were performed with 150 g batches of sucrose. There were three steps of sample preparation. In the first step, cardamom oleoresin (180 mg, 0.12% of sucrose) was accurately weighted in a 50 ml beaker (A). Gum acacia (3.75 g, 2.5% of sucrose) was accurately weighted in another 50 ml beaker (B). Distilled water (9.5 ml) was added to B to make 40% gum acacia solution with continuous stirring with a glass rod until it dissolved. The gum acacia solution was allowed to stand at 25 °C for 1 h for complete hydration of the gum. In the second step, the solutions in A and B were mixed using a shear homogenizer and kept undisturbed to form a stable emulsion (C). In the third and final step, sucrose syrup of 75 °brix was concentrated by heating in a metallic vessel on a heating mantle with continuous stirring until it reached above 95 °brix. The beginning of crystallization was detected as slight sand like turbidity in the syrup. At this point, gum acacia emulsified cardamom oleoresin (C) was added rapidly to the syrup, and heating was controlled while being continuously stirred. The temperature was monitored with a thermocouple. The maximum temperature attained in the process was 130 ± 3 °C. The semi-solid product was then poured into trays and molded into of small cubes of 3.38 cm^3 at 30 ± 2 °C. It was then vacuum dried at 60 °C. The final product was stored in a dessicator. A co-crystallized sample of sucrose without active compound and crystal sucrose were taken as controls. A co-crystallized sample of sucrose without active compound and crystal sucrose were taken as controls.

2.3. Product characterization

2.3.1. Water activity, dissolution time and hygroscopicity

Water activity (a_w) of crystal sucrose and co-crystallized products was determined using Aqua Lab Series 3 TE (USA) equipment. Dissolution time was determined by manually adding crystal sucrose and co-crystallized sucrose cubes (10 g) to 100 ml distilled water with continuous stirring at 400 rpm and 25 °C (Beristain et al., 1994). Hygroscopicity (HG) was calculated from the equation given by (Jaya and Das, 2004) as water gained by the sample on a dry basis as follows:

$$HG\% = \frac{b+H}{a-H} \times 100$$

where b (g) is the weight increase, a (g) is the initial sample weight and H is the initial water content of the sucrose cubes (g). Initial water content (H) was determined by drying the ground co-crystallized sucrose cubes samples in a vacuum oven at 60 °C, until constant weight.

2.3.2. Sorption isotherms

Sorption isotherms were obtained equilibrating 10 g of sample in glass petridish at 25 °C and different relative humidities (RH) within the range 33–93%. Saturated salt solutions used were magnesium chloride (RH 33%), sodium nitrite (RH 63%), and potassium nitrite (RH 93%) (Karel et al., 1996). Sorption isotherms of pure crystal sucrose, co-crystallized sucrose cubes with and without gum acacia emulsified cardamom oleoresin were determined.

2.3.3. Scanning electron microscopy (SEM)

Samples were analyzed in a scanning electron microscope (SEM, Jeol, JSM-6380LA). Co-crystallites were attached to SEM stubs using a two-sided adhesive tape, and then coated with a layer of gold (40–50 nm) and examined using an acceleration voltage of 10 kV and 15 kV for pure crystal sucrose and co-crystallized sucrose samples, respectively.

2.3.4. Differential scanning calorimetry (DSC)

Thermal behavior was studied by using a differential scanning calorimeter (DSC–60), thermal analyzer (TA–60WS), flow control (FC–60A) (Shimadzu, Japan). Crystal sugar and co-crystallized sucrose samples were cooled to 40 °C below the expected glass transition temperature (T_g) and scanned in hermetically sealed pans under a nitrogen flow of 10 ml/min used to purge the sample head and glove box. Heat flow and temperature calibration was performed with indium. The melting temperatures (T_m) and enthalpy (ΔH) of melting of pure crystal sucrose, co-crystallized sucrose cubes with and without cardamom oleoresin were determined at a scan rate of 10 °C/min with heating scan up to 250 °C using 4 mg of sample in hermetically sealed aluminum pan.

2.3.5. X-ray diffraction (XRD)

The equipment used was an X'Pert PRO (Holland) at 40 kV with radiation of wavelength of 40 mÅ. Samples were scanned with 2θ between 5 and 60 degree. Co-crystallized products were dried at 60 °C in a vacuum oven before the assay. X-ray diffraction of pure crystal sucrose and co-crystallized sucrose cubes with and without cardamom oleoresin were also determined.

2.3.6. Quantification of active compounds and encapsulation efficiency

To evaluate the encapsulation efficiency or process yield of these encapsulated active components, the co-crystallized sugar cubes were subjected to analysis for entrapped 1,8-cineole and a-terpinyl acetate by method given by (Guenther, 1975; Marriott et al., 2001) with slight modifications. Co-crystallized flavour sucrose cubes were dissolved (25 g) in distilled water (30 ml) in a