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Characterization and storage stability of chlorophylls microencapsulated in different combination of gum Arabic and maltodextrin

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ABSTRACT

Detailed investigations on the physicochemical and structural characterization of chlorophyll loaded microcapsules and their storage stability have not previously been conducted. Therefore, our objective was to encapsulate unstable chlorophylls using different blends of gum Arabic (GA) and maltodextrin (MD) (GA-MD ratios of 5:5, 3:7, and 0:10) by spray-drying to improve storage stability of chlorophylls. An increase in concentration of MD in wall materials was associated with lower moisture content (0.56%), higher encapsulation efficiency (77.19%), chlorophyll content (46.78 μg/g dry powder), degree of crystallinity, and thermal stability of microcapsules. Furthermore, FTIR, XRD, and DSC analyses confirmed inclusion of chlorophylls within microcapsules. The entrapment of chlorophylls within microcapsules enhanced their storage stability at all temperatures (4, 20, and 40 °C) for ten days; notably, microcapsules coated with MD alone showed the highest storage stability (94.7–97.5%). In conclusion, microencapsulation of chlorophylls using MD alone was optimal for enhancing chlorophylls' storage stability.

1. Introduction

There is a worldwide trend to substitute synthetic colorants with natural pigments in food products owing to consumers' concerns about the harmful effects of synthetic colorants. Chlorophylls are widely distributed in green fruits and vegetables, and can be used as potential alternatives to synthetic colorants because they have a brilliant green color, as well as numerous biological activities. Moreover, chlorophylls are known to possess therapeutic properties, including anti-oxidant, anti-inflammatory, anti-bacterial, anti-carcinogenic, deodorizing, and wound healing activities (Hosikian, Lim, Halim, & Danquah, 2010). Despite all these health benefits, there are limitations to the commercial-scale application of chlorophylls as natural colorants because they are very susceptible to environmental stresses, such as oxygen, enzymes, light, high temperature, and acidic or alkaline pH, which result in degradation and discoloration of chlorophylls (Schoefs, 2002; Marquez & Sinnecker, 2008).

In recent years, the interest in microencapsulation of susceptible compounds in a stable wall matrix as a means of protecting functional compounds from environmental conditions (like oxygen, pH, ionic strength, and temperature) and improving the bioavailability has increased (Pourashouri et al., 2014). Microcapsules are typically comprised of core (active) and wall (carrier) materials, and there are various microencapsulation techniques, such as coacervation, fluidization, lyophilization, extrusion, and spray-drying. Among these techniques, spray-drying is regarded as an economical and effective process in the food industry and, practically, it is the most commonly used technique used for microencapsulation of natural components (Edris, Kalemba, Adamiec, & Piątkowski, 2016; Wilkowska, Czyżowska, Ambroziak, & Adamiec, 2017).

The choice of suitable biopolymers as wall materials is critical to the success of microencapsulation by spray-drying because the type of wall material determines the physicochemical and morphological properties of the produced microcapsules. Moreover, it affects the encapsulation efficiency, shelf-life, and degree of protection of sensitive core materials. In previous studies, different types of wall materials have been studied for microencapsulation, including polysaccharides (modified and hydrolyzed starches, cellulose derivatives, and gums), proteins (caseinates, whey proteins, and gelatins), and lipids (mono- and diglycerides and stearic acids) (Mahdavi, Jafari, Assadpoor, & Dehnad, 2016). Among the numerous biopolymers, maltodextrin (MD), with different dextrose equivalents, and gum Arabic (GA) are the most popular and common wall materials for encapsulating active compounds. GA, the exudate polysaccharides from acacia, has been used as a wall material for spray-drying microencapsulation due to its ability to produce a low viscosity solution at high concentrations compared to

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other gums, form a very stable emulsion, and retain volatiles (McNamee, O'Riorda, & O'Sullivan, 2001; Hosseini, Jafari, Mirzaei, Asghari, & Akhavan, 2015). However, in recent years, the application of GA as a wall material has been curtailed by its high price and impurities (Jafari, Assadpoor, He, & Bhandari, 2008). Thus, researchers are trying to use GA in combination with other wall materials or completely replace it with suitable and novel biopolymers. On this basis, starch and its derivatives, such as MD, have been proven to be adequate biopolymers for use with GA. In general, the partially hydrolyzed starch, MD, is utilized as a secondary wall material in spray-drying microencapsulation, offering several advantages such as comparatively low cost, high levels of hygroscopicity and solubility, low viscosity at high solid concentrations, film forming ability, and, finally, good protection against oxidation (Janiszewska-Turak et al., 2017; Moser et al., 2017). However, the greatest limitation of MD is its low emulsifying capacity, and it is thus generally mixed with other wall materials that can form stable emulsions, such as GA.

Some effort has been made to produce microcapsules for chlorophyll and its derivatives by means of spray-drying and to determine the optimal preparation conditions. Specifically, Porrarud & Pranee (2010) noted that the use of OSA-modified starch as a carrier for Zn-pheophytins resulted in the highest chlorophyll content and the longest half-life, when compared to the use of GA and MD. In the case of chlorophyllide, powders produced using GA and MD were more adequate for protecting chlorophyllide from environmental conditions during storage compared to those prepared with soybean protein isolate (Comunian et al., 2011). Recently, the microencapsulation of green pigment extracted from alfalfa by freeze-drying and its application in heated food (jelly and gummy candy) were evaluated by Raei, Yasini Ardakani, & Daneshi (2017). In that study, the authors used a blend of agar and gelatin as wall materials and reported the optimized formula for making microcapsules with the highest encapsulation efficiency. Furthermore, they reported that during heating, the microencapsulated green pigment showed a lower degree of change in the color factors (L^*, a^*, and b^*) than the non-encapsulated green pigment.

However, as far as the present authors are aware, there are no documented detailed investigations on the physicochemical and structural characterization of microcapsules entrapping natural chlorophylls and their storage stability. In our previous study (Kang, Park, Jung, & Chang, 2018), we prepared semi-purified chlorophylls from spinach and analyzed their structure by Fourier transform infrared (FTIR) and nuclear magnetic resonance (NMR) spectroscopy to verify the existence of chlorophylls. Because extracted chlorophylls are easily degraded during storage and processing, we postulated that microencapsulation of chlorophylls in a suitable matrix could provide good protection against adverse environmental conditions. Therefore, the present study was aimed at the development of chlorophyll loaded microcapsules using different blends of GA and MD as protective carriers by spray-drying. To characterize and compare the obtained microcapsules, we evaluated the moisture content, percentage encapsulation efficiency, chlorophyll content, particle size distribution, visual color value, and surface properties of chlorophyll loaded microcapsules. Moreover, characterization of microcapsules was carried out by FTIR spectroscopy, X-ray diffraction (XRD), and differential scanning calorimetry (DSC), and the storage stability of these species at different storage temperatures was evaluated.

2. Material and methods

2.1. Materials

Freeze-dried chlorophylls, as a core material, were prepared according to our previous study (Kang et al., 2018) and kept at −80 °C until use. Bergabest MCT oil 60/40 (Sternchemie GmbH & Co. KG, Germany) was used as a solvent to dissolve the chlorophylls. Maltodextrin (MD; DE = 14–20) (Daesang Co., Seoul, Korea), gum Arabic (GA; Samchun Pure Chemical Co., Seoul, Korea), and Tween® 80 (polyoxyethylene sorbitan monooleate, Junsei Pure Chemical Co., Tokyo, Japan) were used as the wall materials and emulsifier, respectively. Potassium bromide (KBr; anhydrous) and petroleum ether were purchased from Sigma Chemical Co. (St. Louis, MO. USA) and Junsei Pure Chemical Co., respectively. Isooctane, isopropyl alcohol (isopropanol), ethyl alcohol (95%, denatured), and acetone were obtained from Daejung Chemicals Co. (Siheung, Korea). All reagents used in the present study were of analytical grade.

2.2. Preparation of emulsions

GA and MD, as wall materials, were mixed according to the compositions listed in Table S1 (Supplementary data) and dissolved in distilled water (30%, w/v) while stirring at 60 °C for 3 h. The solutions were then cooled to room temperature (23 ± 1 °C) and kept overnight in a refrigerator at 4 ± 0.5 °C to ensure complete hydration of the polymer molecules. After hydration, 1.5% (w/v) of the emulsifier, Tween® 80, was added to the hydrated wall material solutions and completely dissolved with vigorous stirring at room temperature. The core material was prepared by dispersing chlorophylls in MCT oil in the dark to a final concentration of 0.01 g/mL. The oil-in-water (O/W) emulsions were produced by blending the wall material solution and core material in a 2:1 (v/v) ratio using a DIAX 600 homogenizer (Heidolph, Kelheim, Germany) at 20,500 rpm for 3 min.

2.3. Preparation of microcapsules by spray-drying

The freshly prepared emulsions were spray-dried using a laboratory scale SD-1000 spray-dryer (Eyela, Tokyo, Japan) under the following operational conditions: inlet air temperature: 145 °C, outlet air temperature: 95 °C, rotary atomizer: 10 × 10 kPa, blower rate: 0.60 m^3/min, and pump speed: 1.50 mL/min. The obtained microcapsules were stored in sealed conical tubes at −20 °C for further analysis.

2.4. Characterization of emulsions

2.4.1. Emulsion stability

The emulsion stability index (ESI) of each sample of liquid emulsion was evaluated by the volumetric method (Chang, Shin, & Lee, 1994). An aliquot of 10 mL of each emulsion was transferred to a measuring cylinder and kept at room temperature for 24 h. The volume of the separated water layer in the bottom was measured after 24 h, and the ESI within a possible range from 0 to 1 was calculated as follows:

$$ESI = \{1-(V_s/V_a)\} \times 100 \quad (1)$$

where V_a represents the volume of added water in the emulsion and V_s is the volume of the separated bottom layer after the desired storage period. A value of 0 indicates poor emulsion stability, whereas a value of 1 indicates high emulsion stability.

2.5. Characterization of microcapsules obtained by spray-drying

2.5.1. Moisture content

The moisture content of each microcapsule was evaluated using the AOAC method (AOAC 2005). One gram of each microcapsule was placed on an aluminium pan and dried at 105 °C for 1 h in a drying oven. For calculation of the moisture content of each microcapsule, the following equation was used:

$$\text{Moisture content}(\%) = (\text{Wet powder weight - Dried powder weight}) / \text{Wet powder weight} \times 100 \quad (2)$$

2.5.2. Microencapsulation efficiency

The microencapsulation efficiency is defined as the ratio of the core