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Role of solids composition on α-relaxation behavior, molecular structure and stability of spray-dried xanthones encapsulation systems around glass transition

Nattiga Silalai [a, *], Tunyaporn Sirilert [a], Yrjö H. Roos [b], Naritchaya Potes [b], Sakamon Devahastin [c]

[a] Department of Food Technology, Faculty of Science, Siam University, 38 Phetkasem Road, Phasicharoen, Bangkok 10160, Thailand
[b] School of Food and Nutritional Sciences, University College Cork, Cork, Ireland
[c] Advanced Food Processing Research Laboratory, Department of Food Engineering, Faculty of Engineering, King Mongkut's University of Technology Thonburi, 126 Pracha u-tid Road, Tungkru, Bangkok 10140, Thailand

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ABSTRACT

Although the glass transition properties and encapsulation efficiency of various biopolymers have been documented, no attempts have been made to relate the α-relaxation behavior, molecular structure and stability of an encapsulation system above the glass transition. In this work, the efficiency of whey protein (W), maltodextrin (M) and their combination (MW) to encapsulate α-mangostin was assessed through the monitoring of the changes in the mechanical property and molecular structure around the glass transition using dynamic-mechanical analysis and Fourier transform infrared spectroscopy, respectively. A dramatic decrease in the storage modulus was observed in the non-encapsulation system (NE). Addition of W and M increased the temperature difference ($T_{storage} - T_{\alpha}$), resulting in a decrease in the α-mangostin degradation rate during storage. Carbonyl group (C—H) vibration of reducing sugars became smaller when W was added, while the spectra of the M and MW systems exhibited sharp peaks. This confirmed better encapsulation with W than with M.

1. Introduction

Mangosteen (*Garcinia mangostana* L.) or "Mang Khut" in Thai is a tropical fruit that has been widely consumed due to its tasty flesh. Mangosteen pericarp has also been used as a traditional medicine for treating such a symptom as diarrhea due to its xanthones, which are a class of phenolic compounds and also one of the most potent natural antioxidants (Ji et al., 2007; Zadernowski et al., 2009). Xanthones are noted to consist of α-mangostin, β-mangostin, 9-hydroxycalabaxanthone, 3-isomangostin, gartanin and 8-deoxygartanin as determined by high-performance liquid chromatography (HPLC). α-mangostin has, in particular, been reported to have the highest antioxidant activity (Ji et al., 2007; Okonogi et al., 2007). Prior to their use, however, xanthones must normally undergo thermal processing, which leads to their degradation. Suvarnakuta et al. (2011), for example, reported the losses of xanthones and their antioxidant activity in mangosteen rind during drying; methods and conditions of drying affected the changes of xanthones and their antioxidant activity. A means to increase the stability of the compounds both during processing and storage is therefore much desired. Among many possible alternatives, encapsulation via spray drying is an effective technique for entrapment of and hence protecting bioactive compounds or sensitive ingredients inside a coating material or a continuous phase (Saenz et al., 2009; Ahmed et al., 2010; Fang and Bhandari, 2010; Sansone et al., 2011).

Encapsulation is normally achieved via the use of a biopolymer that can help improve product stability by forming a solid, amorphous continuous phase (glass) surrounding a target compound to be protected. An array of biopolymers, including various proteins, maltodextrin and cyclodextrin have been used either alone or in combination with other materials to encapsulate plant extracts, aromatic additives, carotenoids and vitamins (Guzey and McClements, 2006; Gunasekaran et al., 2007; Bae and Lee, 2008;

* Corresponding author.
E-mail address: nattiga.silalai@gmail.com (N. Silalai).

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Saenz et al., 2009; Livney, 2010; Sansone et al., 2011). Different types of encapsulating materials clearly result in different molecular structures and mobilities of amorphous solids as well as encapsulation systems that form during spray drying. It is indeed known that proteins, polysaccharides and their blends can improve the functional properties of food formulations (Benichou et al., 2002). For example, Shaikh et al. (2006) indicated that polysaccharides such as mesquite gum (MG) possess emulsifying and microencapsulating properties, forming dense films upon drying that limit oxygen diffusion through its matrix. On the other hand, maltodextrin exhibits multiple capabilities, including an ability to bind sensitive compounds as well as to reduce the oxygen permeability of the wall matrix (Bae and Lee, 2008). Use of a mixture of maltodextrin and pectin led to the formation of stable powders and a capability to mask unpleasant smell of a product (Sansone et al., 2011). Whey protein also serves as a good encapsulating material over a wide range of pH of food applications (Morr and Ha, 1993).

The characteristics of spray-dried materials are often governed by glass transition, which in turn depends on the molecular weight and miscibility of the various components of the solids (Roos and Silalai, 2011). Ahmed et al. (2010), for example, revealed that maltodextrin could protect such encapsulated ingredients as ascorbic acid from oxidation by increasing the overall glass transition temperature (T_g) of the encapsulation system as a result of improved miscibility of the mixtures. On the other hand, protein was noted not to affect the T_g but form thin films, which could help improve the glass-forming ability of particle surfaces during spray drying (Fang and Bhandari, 2012). Ubblink and Krüger (2006), among others, indicated the glass formation of amorphous matrices during the encapsulation process and emphasized the importance of the glass-forming ability of biopolymer components in protecting bioactive compounds or sensitive ingredients. Measurement of the T_g is therefore of importance because the encapsulation efficiency and physicochemical properties of an encapsulation system are linked to such a temperature (Silalai and Roos, 2011a; Roos and Silalai, 2011).

Above the T_g powder stickiness and loss of the structural integrity of an encapsulation system are generally observed due to an increase in the molecular mobility of encapsulating materials; this in turn leads to the loss or degradation of the encapsulated bioactive compounds (Flink and Karel, 1970; Ubblink and Krüger, 2006; Bae and Lee, 2008; Roos, 2010). Molecular mobility of encapsulating materials should therefore be assessed. Among the possible evaluation alternatives, Fourier transform infrared (FTIR) spectroscopy and dynamic-mechanical analysis (DMA) can be used (Cruz et al., 2001; Adina et al., 2010). Around the glass transition, the so-called α-relaxation occurs when molecules gain transitional mobility and long-range molecular motions. Amorphous materials exhibit increased molecular mobility, time-dependent changes and rapidly decreasing viscosity above the glass transition, which is responsible for the liquid-like behavior, viscous flow and α-relaxation behavior (Roudaut et al., 2004). α-relaxation in fact shows an onset of long-range molecular motions and can be located through a change in the complex modulus occurring at the α-relaxation temperature (Silalai and Roos, 2011a).

Since changes in the molecular structure are expected to reflect in the changes of the mechanical property and encapsulation efficiency of an encapsulation system, the purpose of this study was to investigate the efficiency of selected biopolymers to encapsulate xanthones as well as to investigate the changes in the α-relaxation and molecular structure of the encapsulation systems. Whey protein and maltodextrin as well as mangosteen pericarp (containing xanthones) powder were used to form the test encapsulation systems. The sugar component, α-mangostin content (as a representative of xanthones), glass transition temperature, molecular structure, morphology and mechanical property were also determined.

2. Materials and methods

2.1. Chemicals and reagents

Xanthones standard (α-mangostin) was purchased from Sigma–Aldrich (St. Louis, MO). Ethanol, methanol and deionized water (HPLC grade) were purchased from Lab-Scan Analytical Sciences (Bangkok, Thailand). Maltodextrin (M; DE ~ 16) was supplied by National Starch and Chemicals (Thailand) Co., Ltd. (Bangkok, Thailand). Whey protein (W) concentrate (WPC) was obtained from Vicchi Enterprise Co., Ltd. (Bangkok, Thailand). Dried powder of mangosteen pericarp was commercially supplied by Thiptipa Co., Ltd. (Bangkok, Thailand).

2.2. Powder production

2.2.1. Liquid feed preparation

A liquid feed with a solid content concentration of 20% (w/w) was prepared from a mixture of mangosteen pericarp powder, suspension of protein phase (WPC) and carbohydrate phase (maltodextrin) at different proportions as listed in Table 1. Suspension of the protein phase was obtained from 15% (w/w) WPC in distilled water; the suspension of the carbohydrate phase was also prepared at 15% (w/w) in distilled water.

For the preparation of M and W encapsulation systems, suspension of each phase was added with 5% (w/w) mangosteen pericarp powder. Suspension of the carbohydrate phase (7.5% w/w) was mixed with the suspension of the protein phase (7.5% w/w) and mangosteen pericarp powder (5% w/w) to prepare the MW encapsulation system. The non-encapsulation (NE) system simply composed of the dry mangosteen pericarp powder; this was prepared by adding 5% (w/w) mangosteen pericarp powder in distilled water without any biopolymers. The mixture of each sample was stirred by an agitator (IKA Labortechnik, Eurostar Basic, Staufen, Germany) at 200 rpm for 3600 s and then sonicated in an ultrasonic sonicator (Lennox, 2645 Ultrawave, Cardiff, UK) at a frequency of 30 kHz for 300 s. The content was then homogenized for 900 s at 11,000 rpm by a homogenizer (IKA Labortechnik, T25-B, Selangor, Malaysia) prior to spray drying.

2.2.2. Spray drying

Each prepared mixture was spray dried via a Büchi B-290 Mini Spray Dryer (Büchi Laboratoriums-Tecnik, Flawil, Switzerland) equipped with a 1.4-mm pressure nozzle under the following experimental conditions: inlet air temperature of 180 °C (453 K), outlet air temperature of 70–80 °C (343–353 K), liquid flow rate of 8.8 mL/min (0.147 mL/s), nozzle pressure of 30 mbar and aspirator opening of 100%. In order to maintain the mixture homogeneity, the suspension was gently stirred via a magnetic stirrer (Heidolph®, MR Hei-Standard, Schwabach, Germany) while being fed into the spray dryer. All spray-dried samples were collected and stored in a hermetically sealed plastic bag at room temperature until further characterization.

2.3. Powder characterization

2.3.1. Dynamic-mechanical analysis (DMA)

Changes in the mechanical property of each encapsulation system were determined using a dynamic-mechanical analyzer (Tritec, 2000 DMA, Triton Technology Ltd., Loughborough, UK).