

Carbohydrate and Protein as Carrier Systems for Encapsulation of Xanthone using Spray Drying

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ABSTRACT: Mangosteen (*Garcinia mangostana* Linn.) pericarp is well known as the natural sources of xanthenes, which contain high antioxidant activity. Drying methods significantly causes loss of xanthenes due to thermal degradation. The retention of bioactive compounds could be improved using encapsulation technique entrapping sensitive ingredients inside the coating material prior to spray drying. In the present study, effect of maltodextrin and whey protein on α -mangostin content, antioxidant activity, physicochemical property of encapsulation systems was determined. A liquid feed concentration (20%, w/w) was prepared by mixing mangosteen pericarp powder, maltodextrin and whey protein prior to spray drying. Non-encapsulation system was prepared using mangosteen pericarp without maltodextrin or whey protein. The results indicated that different types of biopolymers used affected the encapsulation efficiency as well as physicochemical properties of encapsulation systems during storage. Spray-dried mangosteen pericarp encapsulated with protein showed higher T_g and lower changes in storage modulus than spray-dried xanthone encapsulated with maltodextrin and non-encapsulated spray-dried system. Antioxidant activity and α -mangostin content also remained constant during storage for spray-dried mangosteen pericarp encapsulated with protein. Protein could hinder the carbonyl group of reducing sugars leading to molecular mobility.

Keyword: xanthone; maltodextrin; whey protein; encapsulation; spray drying

INTRODUCTION

Mangosteen (*Garcinia mangostana* L.) pericarp has indeed been used as a traditional medicine for such a symptom as diarrhea due to its xanthenes, which are a class of phenolic compounds and also one of the most potent natural antioxidants [1,2]. The 6 xanthone derivatives are mostly found in mangosteen pericarp as α -mangostin, β -mangostin, 9-hydroxycalabaxanthone, 3-isomangostin, gartanin and 8-deoxygartanin. The α -

mangostin has been reported to have the highest antioxidant activity [1,3]. The losses of xanthenes and their antioxidant activity in mangosteen rind during drying; methods and conditions of drying affected the changes of xanthenes and their antioxidant activity [4]. Wang et al. [5] indicated that degradation or oxidation process taking place during storage period could reduce the polyphenolic content and consequently the nutritional value. A means to increase the stability of the compounds both during processing and storage is therefore much desirable. Encapsulation via



spray drying is an effective technique for entrapment of bioactive compounds or sensitive ingredients inside a coating material or a continuous phase before spray drying [6]. Biopolymers are encapsulants that improve product stability by forming a solid, amorphous continuous phase (glass) due to the removal water. Ubblink and Krüger [7] have discussed the glass formation of amorphous matrices during the encapsulation process and emphasized the importance of the glass-forming ability of food components in protecting sensitive ingredients. Physicochemical properties and the efficiency of encapsulation are well known to be linked to the glass transition temperature [8]. Above glass transition (T_g), disruption of the structural integrity of wall matrix or particle surface occurred due to an increase in molecular mobility of amorphous solids of encapsulating materials causing loss of bioactive ingredients [7,9,10]. Different types of encapsulating materials may result in molecular structure and mobility of amorphous solids as well as encapsulation system via spray drying method. Ahmed et al. [11] revealed that maltodextrin could protect encapsulated ingredient, such as ascorbic acid from oxidation and increase overall T_g values due to the increase of molecular weight of the components, while protein did not affect the T_g values but it formed a thin protein-film as well as improved glass-forming of particle surfaces during spray drying [12]. Spray drying was negatively affected by increasing level of sugar, which decreased the measured glass transition temperature (T_g) of the dehydrated foods in agreement with the reported difficulty of spray drying sugar-rich products with low T_g as a result of plasticized particle surfaces [13]. The addition of substances with high T_g values (i.e., protein and maltodextrin) into a blend prior to spray drying increased T_g of mixed system as well as reduced molecular mobility of low molecular weight solids in the spray-dried system [14]. Although the glass transition properties of encapsulated systems are well documented, the compositional effect

and molecular mobility of amorphous solids on encapsulation efficiency have not been reported. The purpose of this study was; therefore, to evaluate the encapsulation efficiency of biopolymers used for the extension of the limited shelf life of oxidized spray-dried xanthenes as well as the changes in physicochemical property and molecular mobility of solids composition around glass transition temperature. In the present study, whey protein, maltodextrin and mangosteen pericarp powder were used as the spray-dried encapsulation systems. The α -mangostin content, radical-scavenging activity, glass transition temperature, molecular structure, mobility, morphology and mechanical property were determined. Interpretation as well as discussion of the significant outcomes of all results has been carried out in this research.

MATERIALS AND METHODS

Materials

Xanthone standard, α -mangostin, purchased from Sigma-Aldrich (Louis, MO). Ethanol, methanol and deionized water (HPLC grade) were purchased from Lab-Scan Analytical Sciences (Bangkok, Thailand). These reagents were analytical chemical grade. Highly esterified pectin (degree esterification 70-75%) and maltodextrins (DE~16) were supplied by National Starch and Chemical (Thailand) Co., Ltd (Bangkok, Thailand). Whey protein concentrate (WPC) was obtained by Vichhi Enterprise Co., Ltd (Bangkok, Thailand). Mangosteen pericarp powder was commercially supplied by Thiptipa Co., Ltd (Bangkok, Thailand). They were food grade substances.

Sample preparation

A liquid feed concentration with the total concentration of 20% (w/w) of each treatment was prepared prior to spray drying. The liquid suspensions were separately prepared into 3 treatments: (i) a mixture of 5% (w/w) mangosteen pericarp solution and 15% (w/w)



maltodextrin solution, (ii) mixture of 5% (w/w) mangosteen pericarp and 15% (w/w) whey protein, and (iii) only 20% (w/w) mangosteen pericarp. The liquid suspension of each treatment was stirred by an agitator at 200 rpm for 30 min, and homogenized (IKA Labortechnik T25-B, Selangor, Malaysia) at 11000 rpm for 15 min prior to spray drying.

Spray drying

Each prepared mixture was spray dried via a Büchi B-290 Mini Spray Dryer (Büchi Laboratoriums-Technik, Flawil, Switzerland) equipped with a 1.4-mm pressure nozzle under the following experimental conditions: inlet air temperature of 180 °C; outlet air temperature of 70-80 °C; liquid flow rate of 8.8 mL/min; pressure of 30 mbar and aspirator 100%. In order to maintain homogeneity, the suspension was gently stirred via a magnetic stirrer (Heidolph Instruments, 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 before further characterization.

Powder characterization

Water sorption: Approximately 1 g of each powder sample was transferred into glass vials and dried in a vacuum oven at temperature of 50 °C for 24 h. Water sorption behavior of the sample was determined according to method of Silalai and Roos [14]. The dried powders were equilibrated in evacuated desiccators over saturated salt solutions of LiCl, CH₃COOK, MgCl₂, K₂CO₃, Mg(NO₃)₂, NaNO₂, NaCl, KCl and K₂SO₄ (Sigma Chemical Co., St. Louis, Mo., U.S.A.) at corresponding a_w of 0.11, 0.23, 0.33, 0.44, 0.54, 0.65, 0.76, 0.84 and 0.97, respectively, at ambient temperature. Water content at each a_w was determined from the mean weight of triplicate samples. Steady-state water contents of the powders at 240 h were obtained for sorption isotherms. The Guggenheim-Anderson-deBoer (GAB) model was fitted to the experimental data.

Glass transition

Glass transition temperature (onset T_g) was determined via the use of a differential scanning calorimeter (DSC 204 F1 Phoenix®, NETZSCH-Gerätebau GmbH, Germany), and analyzed via NETZSCH Proteus® software, version 6.1 (NETZSCH-Gerätebau GmbH, Germany). Glass transition temperature was determined according to method of Silalai and Roos [14] with some modification. A spray-dried sample (1 g) was transferred to a glass vial and equilibrated in an evacuated desiccator over P₂O₅ and saturated salt solutions of LiCl, CH₃COOK, MgCl₂ and K₂CO₃. The equilibrated sample (5-10 mg) was then in a DSC aluminum pan (40 µL; NETZSCH-Gerätebau GmbH, Germany) and hermitically sealed. An empty pan was used as a reference. The sample was scanned first to 40 °C above the predetermined T_g (onset) at 5 °C/min, then cooled at 10 °C/min to 40 °C below the T_g (onset); and the second heating scan at 5 °C/min was run to well above the glass transition temperature range. Anhydrous sample was scanned using a pan with a punctured lid to allow evaporation of residual water during the measurement. The Gordon-Talyor (G-T) model was fitted to the experimental T_g data. Triplicate samples of each powder were analyzed.

Mechanical property

Changes in the mechanical property of the encapsulation systems were determined using a dynamic-mechanical analyzer (Tritec 2000 DMA version 1.43.00 software, Triton Technology Ltd., Loughborough, UK). Approximately 60 mg of each equilibrated sample was spread on a metal pocket-forming sheet (Triton Technology Ltd., Loughborough, UK) according to the method of Silalai and Roos [8]. The powder was analyzed dynamically at a heating rate of 3 °C/min from approximately 40 °C below to approximately 40 °C above the observed onset α -relaxation temperature (T_α). During the dynamic heating, the sample was analyzed for E' , E'' and $\tan \delta$



at frequencies of 0.5, 1 and 5 Hz. The Onset temperature of α -relaxation was taken as the temperature at a drop in the storage modulus. Changes in mechanical property around the glass transition were also observed from the α -relaxation behavior.

Quantitative analysis

Determination of sugar compositions: Sugar contents (mono- and disaccharide compositions) of a sample were determined using high liquid performance liquid chromatography (HPLC) according to the method of Silalai and Roos [14] with some modifications. A chromatographer (Agilent Technologies, LC1200 HPLC, Waldbronn, Germany) with diode array and multiple wavelength detectors (Agilent 1200 Series, Waldbronn, Germany) was used. The eluent was 20 mM NaOH prepared from 50% (w/w) HPLC-grade NaOH (Sigma-Aldrich, Steinheim, Germany) and sonicated (Lennox 2645, Ultrawave Limited, Cardiff, U.K.) deionized water. Aliquot was dispensed and filtered using a syringe and 0.45 μ m nylon membrane filters (Millipore Corp., Bedford, Mass., USA.). A standard curve was prepared with 5 different concentrations (10, 30, 50, 70, 100 ppm) of glucose, fructose, maltose and sucrose. At all times during the analysis, the temperature of the detector was maintained at 30 °C.

Determination of α -mangostin content: The α -mangostin content of a sample was determined using HPLC according to method of Suvarnakuta et al. [4] with some modifications. The HPLC system consists of a pump and controller (Waters, model 600, Milford, MA) as well as an absorbance detector (Waters, model 486, Milford, MA). Symmetry® C₁₈ 5 μ m (3.9x150 mm) (Waters, Milford, MA) was used for the analysis of α -mangostin. Mangosteen pericarp powder (0.2 g) was mixed with 3 ml of 95% (v/v) ethanol and extracted in an ultrasonic bath (Lennox 2645, Ultrawave Limited, Cardiff, U.K.)

generating frequency of 30 kHz for 10 min at room temperature. The mixture was then centrifuged (ALC International Srl, model PK-131R, Italy) at 3000 rpm for 5 min. A supernatant was collected and transferred to a 10-ml volumetric flask, and the extract was then filled up to the final volume of 10 ml with 95% (v/v) ethanol. Prior to injection (10 μ l) the ethanolic extract was filtered through a 0.45 μ m nylon membrane filter (Millipore Corp., Bedford, Mass., USA.); the mobile phases were degassed by an ultrasonic bath with the frequency of 30 kHz for 15 min at room temperature. The α -mangostin evaluation assay was performed using a standard curve with different concentrations (0, 25, 50, 75 and 100 mg/ml).

Antioxidant activity

Measurement of antioxidant activity: Antioxidant capacity was determined by the DPPH (2,2-Diphenyl-2-picrylhydrazyl) assay and ABTS (2,2'-Azinobis (3-ethylbenzothiazoline-6-sulphonic acid)) assay according to the method of Masuda et al. [15] and Hiranvarachat et al. [16] with some modifications. Trolox was used as an antioxidant standard. Standard curves for both assays were obtained by measuring the ABTS^{•+} and DPPH[•] scavenging activities of 10, 50, 100, 150 and 200 μ M trolox/mL. The ABTS^{•+} and DPPH[•] scavenging activities (antioxidant capacity) of food extracts were expressed on the dry weight basis as mM trolox equivalent (TE)/100 g sample. Each sample was measured in triplicate. Mean and standard deviation (n = 3) were calculated.

Storage stability of spray-dried xanthone encapsulation system: Evaluation of storage stability of a sample was performed according to the method of Sansone et al. [17] with some modifications. The samples (around 1 g) were placed in glass vials and stored in a container over saturated salt solution of 0.54 a_w at 40 °C for 30 days. At the given times (0, 7, 15 and 30 days), all samples were collected to determine



antioxidant capacity using DPPH and ABTS method.

Statistical analysis

All data were analyzed using the analysis of variance (ANOVA) and presented as mean values with standard deviations. Differences between mean values were established using Duncan's multiple range test; values were considered at a confidence level of 95%. Statistical analysis was conducted using SPSS (version 10.0 statistics software, SPSS Inc., Chicago, IL) to perform all statistical calculations. Each experiment was performed in triplicate.

RESULTS AND DISCUSSION

Effect of solids composition on sorption and crystallization behavior

Water sorption characteristics and most other interactions of food solids with water can be defined by the composition of non-fat food solid such as mainly carbohydrates and proteins. Sorption properties are significantly affected by time-dependent phenomena due to structural transformation and phase transition causing deteriorative changes in low-moisture foods and frozen foods [18]. Several sorption isotherms were determined and presented. In the present study, the sample of spray-dried mangosteen pericarp encapsulated with maltodextrin (ME), with whey protein (WE) and non-encapsulation (NE). Sorption isotherms illustrated steady-state amount of water held by the food solids as a function of a_w or storage humidity at constant temperature [18]. The steady-state water contents over the a_w range used in GAB modeling were those at 240 h, although steady-state values were achieved after 24 to 72 h depending on a_w and material types. In the present study, the GAB isotherm model fitted to water sorption data over the experimental a_w range. The data used in modeling covered the a_w range over which sugar crystallization was unlikely. Water sorption behavior of encapsulation and non-encapsulation at various water activities is

shown in Figure 1.

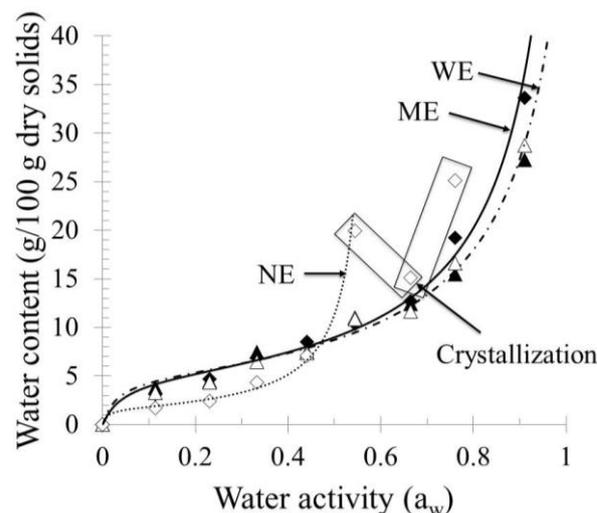


Figure 1 Water sorption behavior of spray-dried mangosteen pericarp with and without encapsulation systems at steady-state water content of storage at ambient temperature (25°C). Values are mean \pm SD (n=3).

Spray-dried mangosteen pericarp without encapsulation (NE) showed substantial differences in their water sorption behavior as compared to the spray-dried mangosteen pericarp encapsulated with maltodextrin (ME) and encapsulated with whey protein (WE). At lower a_w of 0.44, sorbed water was higher in ME and WE than in NE. The NE showed very little moisture gained until the a_w went above 0.44, where sorbed water increased dramatically. This is probably due to hygroscopic behavior of amorphous sugars, which have a strong tendency to sorb the surrounding water using hydrogen-bonds between -OH groups of sugar and water molecules [19]. However, it seemed that sugar crystallization was found in NE above 0.545 a_w as observed in discontinuity of sorption isotherm in the water sorption experiment as a result of loss of absorbed water. Sugar crystallization was achieved when sugar form anhydrous crystals and release all water [14,20]. However, the water released after crystallization was increased and retained to 25% (w/w) in NE. The final water content



after sugar crystallization depended on the types of sugar structure and the other solid components. This was probably due to the resorption of polysaccharide fraction in mangosteen pericarp as well as types and quantities of sugar in spray-dried encapsulation systems. High amounts of sugars (fructose, glucose, sucrose and maltose)

were found in the NE system (Table 1). Sugars with hydrated crystals such as trehalose dehydrates and raffinose trihydrate) can retain higher amounts of water, while sugars with anhydrous crystals such as sucrose and lactose release all water after crystallization [20].

Table 1 Relative amounts of sugars in the carbohydrate fraction of spray-dried mangosteen pericarp non-encapsulated and encapsulated with different biopolymers as analyzed by high performance liquid chromatography (HPLC).

Sugar	Relative amounts of sugars in carbohydrate fraction of spray-dried mangosteen pericarp with and without encapsulation (%)		
	ME	WE	NE
Total sugar	8.41±0.01 ^b	6.66±0.02 ^c	23.63±0.01 ^a
Fructose	1.07±0.01 ^c	1.50±0.01 ^b	12.08±0.01 ^a
Glucose	2.45±0.01 ^c	1.92±0.01 ^b	5.03±0.01 ^a
Sucrose	2.44±0.02 ^b	1.04±0.02 ^c	3.02±0.01 ^a
Maltose	2.45±0.01 ^b	2.20±0.02 ^c	3.50±0.02 ^a

^{a-c} Mean±SD (n = 3) with different superscript alphabets in each row are significantly different (p<0.05).

The ME and WE did not show any decreases in the amount of sorbed water over the a_w range. This suggested that biopolymers such proteins and carbohydrates exhibited interactions with sugars and reduce sugar crystallization either through sugar-protein hydrogen bonding or physically by reducing diffusion or both. A quantity of sugar molecules could be hydrogen bonded to protein molecules and therefore exhibit reduced molecular mobility and rate of crystallization [21]. Moreover, the crystallization process involves diffusion of sugar molecules to crystallization sites [22], which could be probably hindered physically by biopolymers. According to the results, it is likely that steady-state water contents of encapsulation systems (ME and WE) differed slightly at higher 0.65 a_w as a result of differences in types and quantities of biopolymers added in the systems.

Plasticizing effect and glass transition property

Glass transitions of spray-dried mangosteen pericarp without and with encapsulation were determined using DSC. According to the results, the non-encapsulation (NE) system showed the lowest T_g values (Figure 2) due to the presence of mono- and disaccharides in encapsulation systems (Table 1). Glass transition temperatures of sugars governed the physical state of a wide range of foods. All sugars exhibit significant water plasticization, which affects the low T_g values and stability of a number of foods. Glass transition temperature of sugars generally depends on the molecular weight. Monosaccharides have lower T_g values than disaccharides, which have lower values than oligosaccharides. However, the T_g value could be affected by residual water in the samples during process and storage as a result of water plasticization [18]. Hence, an amorphous phase of sugars, especially monosaccharides and the low quantity of



amorphous components in its total solids could be explained the resultant lower T_g in NE system and higher T_g in ME and WE systems. This was consistent with the findings of several studies [11,17] indicating increased T_g by adding some biopolymers such as proteins and carbohydrates in spray-dried systems.

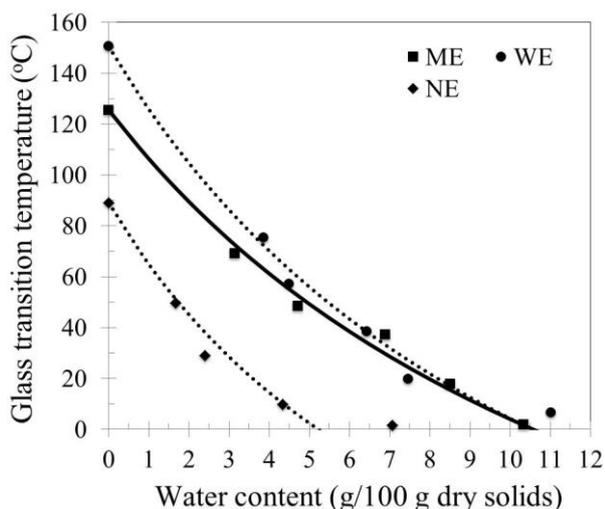


Figure 2 Effects of water plasticization on the glass transition temperatures (T_g , °C) of spray-dried encapsulation and non-encapsulation systems. The T_g values decreased dramatically with increasing water content.

The quality of dehydrated foods could be improved by remaining the material temperatures below the glass transition temperature, which reduces the structural damage and loss of sensitive ingredients of encapsulated powder [9]. In addition, low glass transition temperature of the system could lead to disruption of the structural integrity of wall matrix causing loss of bioactive compounds [7]. Therefore, the addition of substances with high T_g values into a blend prior to spray drying increased T_g of mixed systems and improved characteristics of powders as well as glass-forming of particle surfaces during the process [12]. The T_g values of WE and ME were the higher than those of NE system over

water content range. Carbohydrate-protein interactions were also suggested by the highest T_g values of the WE due to the presence of high amounts of protein, which is typical of high molecular weight components [14]. In addition, the T_g of all systems decreased with increasing water content due to the water plasticizing effect (Roos, 1993). The decreased T_g of all encapsulation systems with increasing water content could suggest that solids components (maltodextrin and whey protein) used may be miscible under certain condition. This was consistent with studies of Kalichevsky and Blashard [23] indicating greater miscibility of biopolymers (proteins and carbohydrates) at high water contents. Although the T_g values of encapsulation systems were closed each other at the lower water contents, their T_g values showed small variations at the higher water contents. Types and quantities of monosaccharide resulted in glass transition behavior of amorphous mixtures of sugars. Arvanitoyannis et al. [24] indicated the plasticizing effect that glucose was plasticized by fructose in anhydrous glasses, which were further plasticized by water.

Mechanical property and molecular mobility

A decrease in storage modulus indicated mechanical α -relaxation, which was related to molecular mobility around the glass transition as described by Silalai and Roos [8]. The effect of biopolymer types on the mechanical property of spray-dried mangosteen pericarp encapsulated systems is shown in Figure 3. Storage modulus showed a small decrease and remained at a high level of the rubbery plateau for the ME and WE around the glass transition temperature, whereas a dramatic decrease in storage modulus was found the in NE. Several studies indicated the correlation of changes in mechanical properties and α -relaxation around the glass transition, which enhanced molecular mobility [24]. A dramatic decrease in storage modulus indicated high molecular



mobility for component molecules in sugar-rich powder. The dominant role of sugar was the main component contributing to changes in modulus around the glass transition [25]. The α -relaxation energies were dependent on the molecular size and to some extent on hydrogen bonding, which was dependent on molecular structure [26]. The results indicated that high amount of sugars in NE resulted in strong thermal plasticization and increased magnitude of a drop in storage modulus as well as depression of T_α relative to the quantity of the low molecular weight substances in the amorphous phase (Table 1). The onset α -relaxation of WE system occurred at the highest temperatures followed by ME system.

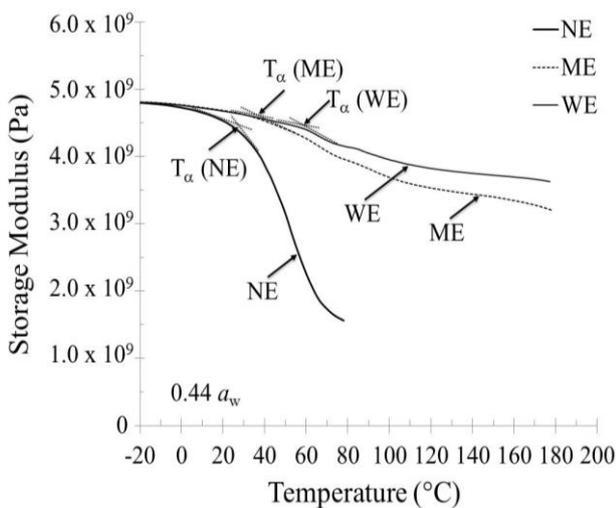


Figure 3 Storage modulus for spray-dried mangosteen pericarp with and without encapsulation with biopolymers at 0.44 a_w . The data were obtained using dynamic heating at 3°C/min at 0.5 Hz. The onset temperature of α -relaxation (T_α) was obtained from the temperature at the drop in storage modulus around glass transition.

In addition, the WE system also showed smaller decrease in storage modulus than the ME system. Proteins are typically high molecular-weight compounds that may interact with sugar molecules and reduce the

number of free sugar molecules available for molecular mobility [21,24]. In addition, Kalichevsky and Blashard [23] also indicated that protein was less plasticized by water and sugars than starch resulting in the smallest decrease in storage modulus for WE in the present study. These findings showed that although sugar was the high component contributing to the mechanical properties, the high molecular weight components such as maltodextrin and protein could reduce overall molecular mobility. This was agreement with the several studies [27] indicating that the presence of biopolymers could reduce the changes in mechanical properties and molecular mobility of sugar-protein mixtures. Changes in mechanical properties were relative to encapsulation efficiency around glass transition, which enhanced molecular mobility of solid components [8,11,17]. Increased molecular mobility may induce crystallization and collapse structure above glass transition leading to loss of bioactive compounds [11,17]. Therefore, bioactive compounds, such as xanthone could be released from the matrices during processing and storage, while the release may decrease below T_g due to glassy structure and rigid molecular mobility.

Changes in antioxidant activity and α -mangostin content of spray-dried mangosteen pericarp encapsulation systems during storage at accelerated condition

In the present study, the antioxidant efficiency of xanthone encapsulated in different types of biopolymers was investigated under accelerated storage condition. The functional stability of encapsulation and non-encapsulation systems was evaluated as free-radical scavenging activity using the DPPH and ABTS tests [4]. The α -mangostin content, a strong and well-known antioxidant of xanthones, was used as positive control assay. All samples contained α -mangostin, which is generally known to react with various components and undergo to oxidation or degradation during process

and storage [3]. During storage at accelerated condition (40 °C and 0.54 a_w), the antioxidant activity determined by DPPH and ABTS method is shown in Figure 4. The DPPH and ABTS scavenging activity of NE system decreased dramatically, whereas those of ME and WE systems decreased slightly during storage. This result was related to the α -mangostin content showing a dramatic decrease in non-encapsulation system but a slight decrease in the encapsulation systems during storage. This suggested the efficiency of whey protein and maltodextrin used for xanthone encapsulation. The present study indicated that α -mangostin degradation was closely related to moisture sorption behavior and changes in mechanical properties around the glass transition.

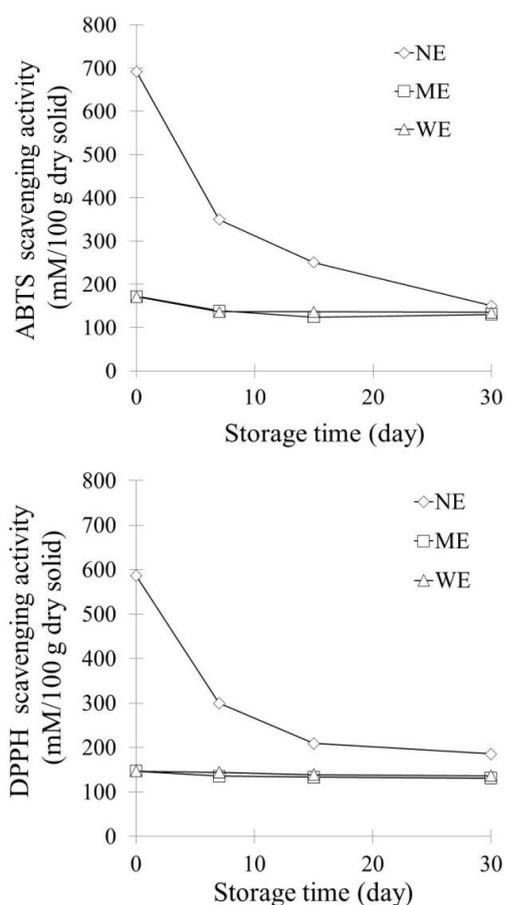


Figure 4 Antioxidant activity of encapsulation and non-encapsulation systems during storage in accelerated conditions for 30 days (0.54 a_w and 40°C).

At storage condition of 0.54 a_w , particle surface of spray-dried NE system was probably completely lost due to plasticizing effect and depression of T_g leading to mobility of reactants and solids, dissolution of wall materials, crystallization and extensive fusing of the particles above glass transition, whereas the particle surfaces of ME and WE system could remain stable in the glassy state. Leung [28] indicated that increased pro-oxidant effect at high a_w was due to the increased mobility of reactants. Water mobility mechanism may take place at higher a_w above 0.54, since water acts as a plasticizer of the carbohydrate wall matrix resulting in viscous flow, enhanced mobility and an eventual structure collapse [10].

CONCLUSION

Encapsulation systems had high values and could remain antioxidant activity during storage. Water sorption and molecular mobility as well as antioxidant activity of encapsulation and non-encapsulation systems were affected by the solids composition and water content. Stability of the α -mangostin was significantly affected by water activity and low molecular weight sugars as a result of their molecular mobility and structure collapse, which could be reduced by the addition of high molecular weight components such as maltodextrin and whey protein. The present study indicated that xanthone degradation can be reduced for encapsulation systems with possible lower molecular mobility and structure collapse of the particle surfaces. Physical properties of amorphous mixtures affected their encapsulation stability of food powders. Therefore, applicability of biopolymers, such as maltodextrin and whey protein could be used as encapsulating agents of sensitive ingredients or bioactive compounds in food products. However, it seemed that whey protein was a better encapsulating agent than maltodextrin at the same condition. Protein reduced the overall molecular mobility, while

maltodextrin increased overall T_g of the encapsulation systems.

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REFERENCES

- [1] Ji, X., Avula, B. and Khan, I.A. (2007). Quantitative and qualitative determination of six xanthenes in *Garcinia mangostana* L. by LC-PDA and LC-ESI-MS. *Journal of Pharmaceutical and Biomedical Analysis*. 43: 1270-1276.
- [2] Zadernowski, R., Czaplicki, S. and Naczka, M. (2009). Phenolic acid profiles of mangosteen fruits (*Garcinia mangostana*). *Food Chemistry*. 112: 685–689.
- [3] Okonogi, S., Duangrat, C., Anuchpreeda, S., Tachakittirungrod, S. and Chowwanapoonpohn, S. (2007). Comparison of antioxidant capacities and cytotoxicities of certain fruit peels. *Food Chemistry*. 103: 839-846.
- [4] Suvarnakuta, P., Chaweerungrat, C. and Devahastin, S. (2011). Effects of drying methods on assay and antioxidant activity of xanthenes in mangosteen rind. *Food Chemistry*. 125: 240-247.
- [5] Wang, Y., Zhaoxin, L., Fengxia, L. and Xiaomei, B. (2009). Study on microencapsulation of curcumin pigments by spray drying. *European Food Research and Technology*. 229: 391-396.
- [6] Saenz, C., Tapia, S., Chavez, J. and Robert, P. (2009). Microencapsulation by spray drying of bioactive compounds from cactus pear (*Opuntia ficus-indica*). *Food Chemistry*. 14: 616-622.
- [7] Ubblink, J. and Krüger, J. (2006). Physical approaches for the delivery of active ingredients in foods. *Trends in Food Science and Technology*. 17: 244-254.
- [8] Silalai, N. and Roos, Y.H. (2010b). Dielectric and mechanical properties around glass transition of milk powders. *Drying Technology*. 28: 1-11.
- [9] Flink, J. and Karel, M. (1970). Effects of process variables on retention of volatiles in freeze-drying. *Journal of Food Science*. 35: 444-446.
- [10] Roos, Y.H. (2010). Glass transition temperature and its relevance in food processing. *Annual Review of Food Science and Technology*. 1: 469-496.
- [11] Ahmed, M., Akter, M.S., Lee, J. and Eun, J. (2010). Encapsulation by spray drying of bioactive components, physicochemical and morphological properties from purple sweet potato. *LWT*. 43: 1307-1312.
- [12] Fang, Z. and Bhandari, B. (2012). Comparing the efficiency of protein and maltodextrin on spray drying of bayberry juice. *Food Research International*. 48: 478-483.



- [13] Bhandari, B.R., Datta, N., Crooks, R., Howes, T. and Rigby, S. (1997). A semi-empirical approach to optimize the quality of drying aids required to spray drying sugar-rich foods. *Drying Technology*. 15: 2509-2525.
- [14] Silalai, N. and Roos, Y.H. (2010a). Roles of water and solids composition in the control of glass transition and stickiness of milk powders. *Journal of Food Science*. 75: E285-E296.
- [15] Masuda, T., Yonemori, S., Oyama, Y., Takeda, Y., Tanaka, T. and Andoh, T. (1999). Evaluation of the antioxidant activity of environmental plants: Activity of the leaf extracts from seashore plants. *Journal of Agricultural and Food Chemistry*. 47: 1749-1754.
- [16] Hiranvarachat, B., Suvarnakuta, P. and Devahastin, S. (2008). Isomerisation kinetics and antioxidant activities of β -carotene in carrots undergoing different drying techniques and conditions. *Food Chemistry*. 107: 1538-1546.
- [17] Sansone, F., Mencherini, T., Picerno, P., d'Amore, M., Aquino, R.P. and Lauro, M.R. (2011). Maltodextrin/pectin microparticles by spray drying as carrier for nutraceutical extracts. *Journal of Food Engineering*. 105: 468-476.
- [18] Roos, Y.H. (1993). Water activity and physical state effects on amorphous food stability. *Journal of Food Processing and Preservation*. 16: 433-447.
- [19] Labuza, T.P. (1968). Sorption phenomena in foods. *Food Technology*. 22: 263-265, 268, 270, 272.
- [20] Buera, P., Schebor, C. and Elizalde, B. (2005). Effects of carbohydrate crystallization on stability of dehydrated foods and ingredient formulations. *Journal of Food Engineering*. 67: 157-165.
- [21] López-Díez, E. and Bone, S. (2000). An investigation of the water-binding properties of protein+sugar systems. *Physics in Medicine and Biology*. 45: 3577-3588.
- [22] Hartel, R.W. (2001). Recrystallization processes. *Crystallization in Foods*. Gaithersburg: Aspen Publishers Inc.
- [23] Kalichevsky M.T. and Blashard J.M.V. (1992). A study of the effect of water on the glass transition of 1:1 mixtures of amylopectin, casein and gluten using DSC and DMTA. *Carbohydrate Polymers*. 19: 271-278.
- [24] Arvanitoyannis, I., Blanshard, J.M.V., Ablett, S., Izzard, M.J. and Lillford, P.J. (1993). Calorimetric study of glass transition occurring in aqueous glucose:fructose solutions. *Journal of the Science of Food and Agriculture*. 63: 177-188.
- [25] Cruz, I.B., Oliveira, J.C. and MacInnes, W.M. (2001). Dynamic mechanical thermal analysis of aqueous sugar solutions containing fructose, glucose, sucrose, maltose and lactose. *International Journal of Food Science and Technology*. 36: 539-550.
- [26] Noel, T.R., Ring, S.G. and Whittam, M.A. (1992). Dielectric relaxations of small carbohydrate molecules in the liquid and glassy states. *Journal of Physical Chemistry*. 96: 5662-5667.
- [27] Silalai, N. and Roos, Y.H. (2011). Mechanical relaxation times as indicators of stickiness in skim milk-maltodextrin solids systems. *Journal of Food Engineering*. 106: 306-317.
- [28] Leung H. (1978). Influence of water activity on chemical reactivity. In L.B. Rockland, L.R. Beuchat, (Eds), *Water activity: Theory and Applications to Food*, IFT Basic Symposium Series. New York: Marcel Dekker Inc., 27-54.

