Microencapsulation of refined liquid smoke using maltodextrin produced from broken rice starch

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Abstract

Many bioactive compounds are contained in liquid smoke, one of which is phenol. Besides, its liquid form causes handling complication during distribution and application, thus the microencapsulation was used to solve it. In the present a various inlet air temperatures were applied (120, 130, 140 ºC) with feed flow rate of 5 mL/minute. Total soluble solid (TSS) of broken rice starch maltodextrin were 20, 25, and 30 %. The observed parameters were moisture content, yield, phenol totals, microencapsulation efficiency,shape and morphology, particle size distribution and volatile compound profile identification. All data was taken triplicate using completely randomized design (CRD). The results showed that the increase of wall material concentration and inlet air temperature were able to improve yield, phenol total, microencapsulation efficiency, and solubility of microcapsules. The best results with highest microencapsulation efficiency of 52.24±2.84 % and 20 minutes phenol release period was obtained using 25 % TSS and inlet temperature of 140 ºC. Volatile

compounds identification using GC/MS showed that the combination between wall material and inlet air temperature was effective to retain phenol as one of liquid smoke bioactive compounds. Microcapsules morphology analysis showed that the average diameter of microcapsules was 4.28 μm and had nearly spherical shape, little shriveled, and without agglomeration with low levels of damage.

Keywords: Refined liquid smoke, broken rice, maltodextrin, spray drying, liquid smoke microcapsules.

Nomenclature

LS : Liquid smoke

IR : Indica Rice

DE : Dextrose Equivalent

FF : Fehling Factor

TSS : Total Soluble Solid

db : Dry basis

wb : Wet basis

1. Introduction

Liquid smoke (LS) is condensate obtained from the condensation of wood smoke produced by smoldering wood chips or sawdust under limited oxygen and constituent pyrolysis of wood such as cellulose, hemicellulose, and lignin (Panagan and Nirwana, 2009; Darmadji, 1996). Refined LS contains active compounds such as phenols, carbonyls and organic acids (Girard, 1992; Maga, 1998; Bratzler et al., 1969), so it can be used as a safe alternative food preservative without carcinogenic (Budijanto et al., 2008) and mutagenic materials (Kažimírová and Jablonická, 1994), as well as anti-bacterial and fungicide materials in certain doses (Pszczola, 1995; Dwiyitno and Rudi, 2006). Moreover, LS can act as an antioxidant (Saloko et al., 2014). However, LS has a disadvantage that is easily damaged during storage and sensitive to environmental factors (light, temperature, oxygen), so the technology that can protect the active compound is needed, one of which is encapsulation technique.

Encapsulation is a process of coating a core material that can be either very small liquid/gas particles with an encapsulant agent so that the core particles have physical and chemical properties as desired (Kim et al., 1996). Encapsulation provides protection of the LS bioactive compounds in an encapsulan at a very small size and potential to change the shape of the liquid to the flour that is stable, so easily applied to the food. In addition, encapsulation can change the liquid product into a powder, making it easier to handle.

The successful of encapsulation process is influenced by the selection of technology and materials as encapsulant agent. Encapsulation by spray drying technique is economical, safe, and applicable (Chan, 2011), therefore it is commonly used in the production of

microcapsules. In the spray drying process, quality and efficiency of the final product is influenced by the operating conditions including inlet air temperature and feed concentration (wall material) (Amerie and Maa, 2006; Chegini and Ghobadian, 2005; Goula and Adamopoulos, 2008; Tonon et al., 2008; Caliskan and Dirim, 2013).

The the appropriate of encapsulant agent will improve the efficiency of micro-encapsulation by protecting the active compounds from heat. However, if the amount of encapsulant agent used is too high can cause delution effect, thus causing a decrease in the chemical content of the material (Şahin-Nadeem et al., 2013; Şahin-Nadeem et al., 2011). Thies (2001) explained that the amount of coating material that is commonly used for commercial microcapsules obtained by spray drying technique is 20–30 %.

Inlet air temperature also affects the resulted microcapsules. Inlet air temperature that is too low can cause low evaporation rate causing formation microcapsules with high density membrane, high moisture content, poor fluidity, and prone to agglomerate. However, if the inlet temperature is high, it can lead to excessive evaporation and lead to cracking of the membrane that triggers premature release formation and a decrease in the amount of encapsulated or core materials (Zakarian and King, 1982 dalam Gharsallaoui et al., 2007).

Maltodextrin is a common ingredient used as a encapsulant/coting agents on microencapsulation process by spray drying technique to increase the capacity of the drying process and lowering the stickiness and agglomeration problems during storage, thus a stable product can be obtained (Anwar and Kunz, 201; Hogan et al., 2001; Shu et al., 2006; Teixeira et al., 2004; Bylaite´ et al., 2001; Shaikh et al., 2006).Commercial maltodextrin can be obtained from three plant sources, such as corn starch, potato, and rice. Indonesia is the

world's third-largest rice producer after China and India. According to FAOSTAT 2014, Indonesian rice production in 2012 reached 69,045,141 mT. In the rice milling process, about 15 % broken rice is resulted (Bhullar and Bhullar, 2013). Rice (Oryza sativa L.) is potential source with 90 % starch content. On the other hand, broken rice is abundant and cheap by-product of rice processing in Indonesia, thus make it highly potential to be used to produce maltodextrin.

Based on those descriptions, the research on the broken rice utilization as material for refined LS encapsulation was done. The appropriate of spray drying operating conditions in this case the inlet temperature and feed concentration (amount of maltodextrin), led to produce microcapsules with good quality and efficiency.

2. Materials and methods

2.1. Materials

LS grade I (food grade) was obtained from PT. Tropica Nucifera Industri with pH 2.5 and phenol total 18.83 % (db). Enzyme α-amylase from *B. licheniformis* (Sigma Chemical Co, St. Louis, USA), chitosan obtained from PT. Biotech Surindo-Cirebon with MW 2183778, DD 87 %, viscosity 44.4 cP, moisture content 7.65 %, and ash content 1.04 %. Broken rice IR 64 obtained from local rice mill in Somberembe Hamlet, Village Slomartari, Kalasan district, Sleman regency, Yogyakarta in February 2014. Other chemicals were Na_2CO_3, $CaCl_2$, folinciocalteau, pure phenol, aquadest, HCl, NaOH, helium and whatman no. 41 and no. 1 filter paper.

2.2. Starch broken rice extraction

Broken rice was processed by wet-milled method as described by Hoe et al. (2013) with

some modifications. Broken rice was soaked in water for 4 hours, then ground. Processing of broken rice into flour was made in a traditional market in Yogyakarta, then dried with a cabinet dryer at 50 °C for 24 h to reach moisture content of ~14 %, then sieved with an 80 mesh sieve.

Starch was extracted by protein alkaline protein extraction method as described by Lim et al. (1991) with modification. 100 g rice flour soaked in 500 mL of 0.15 M NaOH for 24 hours. The slurry was then stirred for 30 minutes at 400 rpm and centrifuged at 2226 g for 20 min. The formed sediment was then dissolved in 500 mL of water and then filtered. The solution was then neutralized with 3 M HCl and centrifuged at 2226 g for 15 min. The formed sediment was washed 3 times with water by centrifugation at 2226 g for 15 min, where dark tailing on top layer was scraped while washed. The obtained starch was dried at 50 °C for 24 h to reach moisture content of ~8.3 %, then ground and sieved with a 60 mesh sieve.

2.3. Broken rice starch maltodextrin production

100 g broken rice starch was dissolved in 400 mL of distilled water and 40 ppm $CaCl_2$ was added as a catalyst, then the pH was adjusted to 6 using 0.1 N HCl solution and then added 0.005 mL α-amylase. Hydrolysis was performed at 87±1 °C, for 15 with agitation. Enzyme was inactivated by cooling down to 27 °C, and pH was adjusted to 3±0.5 using 0.1 N HCl. Suspension was incubated for 30 minutes then added 0.1 N NaOH to reach pH ± 6. The obtained dark solution was centrifuged for 30 minutes at 2226 g. The upper clear layer was then poured slowly on whatman no. 41 filter paper that had been installed on the vacuum pump. The clear solution was then dried at 50 °C for 3 days to reach moisture content of

~3.9 %, then ground and sieved with a 60 mesh sieve to obtain maltodextrin.

2.4. Microparticle preparation

Wall material used was maltodextrin obtained from previous process at total solible solid (TSS) of 20%, 25% and 30%, each combined with 1% w/w chitosan. Based on research orientation, the amount of maltodextrin and chitosan added in refined LS solution to reach TSS 20, 25, and 30 % can be seen in Table 1. Wall material was dissolved into 100 mL refined LS, stirred using magnetic stirrer (400 rpm, 30 minutes, room temperature), and homogenized using homogenizer (Ultraturrax T50 Basic IKA Werke, Germany) at 4000 rpm for 2 minutes.Mixture was then filtered by paper filter to separate insoluble precipitate. Microparticle liquid was stored in cool room to prevent oxydation.

2.5. Spray drying

Microcapsule of refined LS was prepared using spray drying method (Lab Plant Sd 05, UK). Inlet air temperature to dry liquid smoke microparticle were 120, 130, and 140 °C, with flow rate of 5 mL/minute, noozle atomizer pressure of 1 bar, and stable drying air volume. Obtained microcapsule powder was placed into air-tight plastic container equipped with silica gel before stored in low humidity cool room for further analysis.

2.6. Analysis methods

2.6.1. Dextrose equivalent

DE of broken rice starch maltodextrin was conducted according Shi *et al*. (2000).

a. Fehling Factor Value

2.5 g of glucose was dissolved in 1000 mL of distilled water. 15 mL solution was taken and added each 5 mL Fehling's solution A and B. Mixture was then boiled and titrated

immediately with glucose solution until the blue color completely disappear and changed to be reddish brown. The amount of titrant was then recorded, and Fehling Factor calculated by :

$$FF \times \frac{\text{Titrant (mL)} \times \text{glucose weight (g)}}{1000} \quad (1)$$

a. DE Value Determination

Maltodextrin 10 g was dissolved in 200 mL of distilled water, then poured into the burette. 50 mL of distilled water was added each 5 mL of Fehling's solution A and B and 15 mL of glucose solution. Mixture was then boiled and titrated immediately with maltodextrin solution until the blue color completely disappear and changed to be reddish brown. The amount of titrant was then recorded, and DE calculated by:

$$DE = FF \times \frac{100}{\text{Maltodextrin solution concentration (g/mL) x titrant (mL)}} \quad (2)$$

2.6.2. Moisture Content

Moisture content of microcapsules and maltodextrin was measured using moisture tester (Ohaus MB 35 halogen, England), due to highly hygroscopic properties of sample by drying as much as 500 mg sample at 150 °C for about 2 minutes till constant weight. Moisture content of refined LS was determined by toluene distillation method according to AOAC, 1970.

2.6.3. Ash content

Ash content of broken rice starch maltodextrin was conducted according to AOAC Official Method 923.03 (AOAC, 1990).

2.6.4. Solubility

Solubility was analyzed according to Loksuwan (2006) method with major modification. 0.5 g sample was mixed with 50 ml of distilled water using a magnetic stirrer at room temperature for 30 minutes then filtered using whatman no.1 filter paper which had previously been dried to constant weight. Filter paper was then dried at 60 °C to constant weigh, then the solubility calculated by its weight. Equation was as follows:

$$\text{Solubility (\%)} = \frac{\text{Sample (g) - Retante (g)}}{\text{Sample (g)}} \times 100 \qquad (3)$$

2.6.5. Total soluble solid

Total Soluble Solid was measured according to AOAC (2005), by dropping sample solution on hand refractometer. TSS was calculated based on the percentage of dissolved solids in solution refined LS microparticle.

2.6.6. Microcapsule yield

Yield was measured by comparing microcapsule weight with amount of total soluble solid/TSS, where TSS value was previously measured based on liquid smoke weight.

2.6.7. Phenol total

Phenol total was analyzed using Senter et al. (1989) method with slight modification. For refined LS, as much as 1 gof refined LS was weighted and dissolved into 100 mL distilled water, then 10 mL taken from it to dissolved again into 100 mL. For refined LS microcapsules, as much as 0.5 g was dissolved into 50 mL, then 5 mL taken from it to dissolved in the 100 mL flask with distilled water (dilution factor = 1000x). 1 mL from each final solutionwas put into tube, added with 5 mL $Na_2CO_3$2 % alkali, incubated for 10 minutes at room temperature, then added with 0.5 mL folin ciocalteau reagen, and shaken

using vortex before 30 minutes incubation in room temperature. Absorbance was measured at 750 nm. Phenol total of refined LS and refined LS microcapsules was calculated using standard curve equation previously obtained.

2.6.8. Microencapsulation efficiency (ME)

Efficiency of refined LS microcapsules was calculated based on phenolic content of refined LS, reflected amount of capsulated phenolic compound. Calculation was made using Cvitanović et al. (2011) equation, which comparing phenol total contained in microcapsule and in initial refined LS (before encapsulation). Phenol weight in microcapsule (% db) was multiplied with the weight of microcapsule obtained. Equation was as follows:

$$\text{ME (\%)} = \frac{\text{Phenol in microcapsusl (g)}}{\text{Phenol in microparticle solution (\% db)}} \times 100 \quad (4)$$

$$\text{Phenol in microparticle solution (\% db)} = \frac{\text{Phenol in LS (\% db)} \times \text{LS Weight (g)}}{\text{Total soluble solid (g)}} \times 100 \quad (5)$$

$$\text{Phenol in micro-capsules (g)} = \text{Phenol total microcapsules (\% db)} \times \text{Microcapsule weight (g)} \quad (6)$$

2.6.9. Phenol release

Phenol Release was conducted according to Loksuwan (2006) solubility method with major modification. Refined LS microcapsules 1 % (w / v) in distilled water was stirred sustainably at 400 rpm. Furthermore, 5 mL solution was taken at minute 1st, 3nd, 6nd, 10nd, 20nd, 30nd and 40ndthen dissolved in the 100 mL flask with distilled water for phenol total analysis measured according to Senter et al. (1989) method.

2.6.10. Microcapsule morphology

Appearance and shape (morphology) of refined LS microcapsules was observed using

Scanning Electron Microscopy (SEM) according to Cerdeira et al. (2005) method, by placing microcapsule samples on aluminum plate using adhesives, coated with thin layer of gold before observation using SEM instrument at 5 kV voltage and 5000 times magnification.

2.6.11. Volatile compound identification

Identification of volatile compounds in microparticle and microcapsules was referred to the method by Chin et al. (2010) with modification. Refined LS microparticle was directly injected into GC-MS, while refined LS microcapsules need to be prepared before injection. As much as 0.25 g of microcapsules were suspended into 10 mL deionized water, shaken using vortex for 30 minutes, and kept inside sealed conical tube prior to GC-MS injection. Analysis conducted using GCMS-QP2010S Shimadzu. Splitl injection mode with injection port temperature of 250°C was used. Column was AGILENT HP 5MS with anlength x internal diameter x thickness of the film = 30 m x 0.25 mm x 0.25 μm. Oven temperature was run gradiently with the settings set to 60°C held 1 min, then raised to 280 °C at a rate of 5°C/min and held for 21 min. Carier gas was helium at a rate of 0.51 mL/min. Detector was MS (Mass Spectrometer) with ionization energy 70 eV scan mass range (m/z) 28–600 with a 250°C detector temperature.

2.7. Experimental design

Completely randomized experiment design was used in this study with two factorials (inlet air temperature and concentration of wall material), where TSS consisted of 20, 25, and 30 % and inlet air temperature consisted of 120, 130 and 140 °C. The differences between the mean values of multiple groups were analyzed by one-way analysis of variance (ANOVA)

with Duncan's multiple methods range tests. ANOVA data with a $p< 0.05$ was classified as statistically significant. SPSS 17.0 statistics and Microsoft Excel 2010 program were used.

3. Result and discussion

3.1. Characterization of maltodextrin broken rice starch

Maltodextrin broken rice starch obtained showed that maltodextrin had characteristics as standard requirements of commercial maltodextrin (Table 2), such as white to yellowish powder or concentrate solution, high solubility, 6 % of maximum water content, pH 4.5–6.5, 0.6 % of maximum ash content, and DE < 20 (Akhilesh et al., 2012; Wang and Wang, 2000; Storz and Steffens, 2004).According to the USA Food and Drug Administration, the maltodextrins are defined a starch hydrolyzate product that is not sweet with DE less than 20 and they are classified as ingredients generally recognized as safe (GRAS) (Marchal et al., 1999).

One of the determinants of the maltodextrins quality is their solubility in water. Good quality maltodextrin has a high solubility index. The result showed that (Table 2) broken rice starch maltodextrin solubility produced was quite high. Wang and Wang (2000), observed that maltodextrin derived from rice starch had short chain. The high solubility of maltodextrin produced allegedly caused by its short linear chain of maltodextrin rice starch and led to lower molecular weight, thus solubility became high. This is supported with Yusraini et al., (2013), who stated that the longer linear chain caused higher molecular weight, so poorly soluble in water.

3.2. Microcapsule yield

One of the factors that can affect the amount of yield produced on microencapsulation using

spray drying technique is the amount of wall material added. The low percentage yield produced can be caused by two possibilities. The first, the least concentration of the added wall materials will cause evaporation of active compounds due to lack of materials that can bind and retain the active compounds from high temperature contact that can degrade their concentration during drying process. The second possibility is the increasing concentration of the wall materials may cause an increase in the viscosity of the microparticle solution. If the viscosity of microparticle solution is high, it will lead to produce microcapsules with high moisture content due to temperature equilibrium during the drying process faster, thus there will be attachment a large number of microcapsules produced in dryer chamber (Tonon et al., 2008; Krishnaiah et al., 2012).

The recent study showed that the increase in yield positively correlated with increasing wall material concentrations used (Table 3) (TPT 30 %), but showed no significant increase ($p>$ 0.05), except in the 140 °C inlet air temperature ($p<$ 0.05) in which the more wall materials used, the yield of produced percentage will be higher.This can be caused by an increase in the total solids in microparticle solution (Şahin-Nadeem et al., 2011; Şahin-Nadeem 2013).

3.3. Microcapsule moisture content

Moisture content of refined LS microcapsules (Table 3) was decreased with the increase of TSS concentration and spray drying inlet temperature,with lowest value obtained at 30 and 20 %TSS and all inlet air temperature($p<$ 0.05). The higher inlet air temperature causes the increase of water mass transfer through surface evaporation resulting microcapsules with low moisture content (Krishnaiah *et al*., 2012). This result was consistent with study by Saloko *et al*. (2012) and Şahin-Nadeem *et al*. (2013).

Lower moisture content was also obtained by increasing wall material concentrations. The addition of maltodextrin on solution prior to drying process will increase TSS and lower moisture content of solution that will be evaporated during the drying process, thus lead to the decrease of water content in the final product (microcapsules). This indicates that the addition of maltodextrin concentrations is able to lower water content in the produced microcapsules (Krishnaiah et al., 2012). This result is in accordance with study by Şahin-Nadeem et al. (2011) which encapsulated mountain tea (*Sideritisstricta*) extract.

3.4. Microcapsule solubility

The use of inlet air temperature on the same TSS showed no significant difference (p> 0.05) (Table 3), except in TSS 30 % with 140 °C inlet air temperature. High inlet air temperature can lead to the possibility to form hard surface layer microcapsules thus lowering microcapsules wettability and reduce the solubility (Goula and Adamopoulos, 2004). Instead, Low inlet air temperature causes microcapsule water content high allowing agglomeration that can accelerate the reconstitution of the microcapsules in the water. The same results have also been reported by Şahin-Nadeem et al. (2013).

Another possibility which can lead to decrease levels of solubility in TSS 30 % is the presence of amylose retrogradation on maltodextrin used. Unhydrolized amylose will form an opaque gel and also affects dissoluble microcapsule particles (Yusraini et al., 2013). Wang and Wang (2000) stated that the maximum concentration of maltodextrin is about 30 %.

3.5. Phenol total

In Table 3, phenol total of refined LS microcapsules decrease in concomitant with the

increase of TSS, due to higher concentration of solid able to diminish phenol content of final products (dilution effect). This result is in agreement with study by (Caliskan and Dirim, 2013; Şahin-Nadeem et al., 2013; Şahin-Nadeem et al., 2011).

Higher inlet air temperature (140 °C with 20 and 25 % TSS) lead to increase phenol total of microcapsules ($p< 0.05$). Higher inlet air temperature causes low water content of the microcapsules, thus phenol concentrated resulting a higher phenol total consequently. However, that was not occured at 30 % TSS, which was caused by the contribution of higher total solids (dilution effect) as mentioned before. Şahin-Nadeem et al. (2011) encapsulated mountain tea extract and reported that there was an increase in phenol totals in the sample by 4 % with the increase in inlet temperature of 145–155 °C, but the increase in inlet temperatures up to 165 °C resulted in a slight decrease in phenol totals. Şahin-Nadeem et al. (2013) encapsulated sage instant and reported an increase in phenol totals at 145–165 °C inlet temperature, but the increase is not significant.Çam et al. (2013) encapsulated phenolicpomegranate peel by 4 types of maltodextrin and showed an increase in phenol content at 130–160 °Cinlet temperature, but did not show a significant increase.

3.6. Phenol release

The recent study (Fig. 1) showed that at inlet air temperatur 120 and 130 °C with 20, 25 and 30 % TSS, the whole phenol in the microcapsules released at minute 10^{th}, 10^{th}, and 3^{th}, while at inlet air temperatur 140 °C with 20, 25 and 30 % TSS, the whole phenol in the microcapsules released at minute 20^{th}, 20^{th}, and 30^{th}. The increase of wall materials used caused the phenol released faster. In addition to be caused by using more maltodextrin (Loksuwan, 2006) that caused higher solubility of microcapsule, thus affected the

accelerated released of phenolic compounds, another cause was the increasing amount of wall materials used caused lower phenol total content (Table 3), so that tme release for releasing phenol in microcapsules entirely became shorter.

Inlet air temperature also affected phenol release of microcapsules. Seen clearly in Fig.1, in the higher inlet air temperature, phenol content of the microcapsules will be greater and it affectes the increasing duration release of phenol compounds. Another thing that plays a role in the phenol release is the ability of chitosan added to bind phenolic compounds (Popa et al., 2000). At 140 °C inlet air temperature with 30 % TSS, microcapsules showed the ability to retard phenol release up to 30 minutes. Hsieh et al., (2006) observed that, by increasing chitosan concentration on microencapsulation of citronella.oil, it was able to increase its time release. Saloko et al. (2014) reported that, combination of maltodextrin-chitosan as material agent in encapsulated LS showed a significant difference in improving phenol total content compared with LS without chitosan.

3.7. Microencapsulation efficiency (phenol)

Interestingly, microcapsules with 25 % TSS showed a significant increase in microencapsulation efficiency compared with 20 % TSS ($p< 0.05$) (Table 3), however at 30 % was not so. This suggested that the use of higher wall materials did not give a significant effect on improving of microencapsulating efficiency. Microencapsulation efficiency can be increased by increasing wall solution solids concentration which can be related to the effect of wall solids concentration on the formation of surface core prior to the formation of crust around the drying droplets (Young et al., 1993 in Gharsallaoui et al., 2007). Similar results have also been investigated by Caliskan and Dirim (2013) which encapsulated sumac

extract using maltodextrin and concluded that the highest efficiency was obtained from the extract containing 25 % TSS. However, using higher TSS (30 %) can reduce efficiency due to the dilution effect as described previously. Çam et al. (2013) conducted a phenolic encapsulation pomegranate peel with 4 types of maltodextrin and showed that the increase in the ratio between maltodextrin with phenolic encapsulated had a degradation effect on microencapsulation efficiency.

Inlet air temperature significantly influence the resulting microencapsulation efficiency. Inlet air temperature releted directly to the drying rate and moisture content of microcapsules. When the air inlet temperature is low, the low evaporation rate causes the formation of microcapsules with high density membranes, high water content, poor fluidity, and easiness of agglomeration. However, a high air inlet temperature causes an excessive evaporation and results in cracks in the membrane inducing subsequent premature release and a degradation of encapsulated ingredient or also a loss of volatiles (Gharsallaoui et al., 2007). Çam et al. (2013), reported that at 160 °C inlet air temperature showed that yield, efficiency, and phenol total of phenolic pomegranate peel microcapsules was higher than at 130, 140, and 150 °C. However, at inlet temperatures above 160 °C showed no significant improving.

3.8. Microcapsule morphology

Analysis was conducted on microcapsule prepared using TSS 25 % and inlet air temperature of 140 °C as best treatment based on highest phenol efficiency. Selected image from the SEM microstructure analysis of the refined LS microcapsule is illustrated in Fig. 2. The SEM result showed that microcapsules had nearly spherical shape, little shriveled, and

without agglomeration with low levels of damage.

Microcapsule morphology of the resulted microcapsules of various microencapsulation of active compounds by using maltodextrin as a single wall material by spray drying technique showed shrinkages on the surface, shriveled, and even had an irregular shape(Tonon et al., 2008; Şahin-Nadeem et al., 2011; Şahin-Nadeem et al., 2013; Caliskan and Dirim, 2013; Saloko et al., 2012).When compared with the results obtained (Fig. 2), it is proved that using combination of maltodextrin-chitosan as material agents gives better result. Chitosan as biofilms can provide a stable layer structure and act as a good barrier, thus able to protect the coated bioactive compounds (Honarkar and Barikani, 2009).

3.9. Volatile compound identification

Analysis was conducted on microparticle solution and microcapsule prepared using TSS 25 % and inlet air temperature of 140 °C as best treatment based on highest phenol efficiency. Fifteen compounds were identified as described in Table 4. There are two compounds with the highest concentration, i.e. acetic acid and phenol. This proves that LS contains components phenol and organic acids in high quantities. Maga (1998) has observed that liquid smoke from wood material composed of 11–92 % water, 0.2–2.9 % phenolic, 2.8–4.5 % organic acid, and 4.6–2.6 % carbonyl. Bratzler et al., (1969), stated that the main component of wood smoke condensate, consisting of carbonyl (24.6 %), carboxylic acid (39.9 %), and phenolic compounds (15.7 %).

After micro-encapsulated refined LS using broken rice starch maltodextrin using 25 % TSS + chitosan 1 % and 140 °C inlet air temperature, 7 volatile compounds were identified, wherein the solution of microparticle previously identified 13 volatile compounds. This

suggests that during the spray drying process involving high heat transfer can cause the loss of some volatile compounds (Nesterenko et al., 2013). In addition, high heat also causes the formation of a new compound in microcapsules. However, a phenolic compound that is the main bioactive component of liquid smoke can be protected with the use of materials which act as a protective coating. (Young et al., 1993 in Gharsallaoui et al., 2007). This proves that maltodektrin as wall material agent can prevent the degradation of the active ingredient (phenol) microcapsules produced (Gustavo and Canovas, 1999).

4. Conclusion

Combination of 25 % broken rice starch maltodextrin and 1 % chitosan (w/w) as wall materials and the applied 140 °C air inlet temperature showed good ability to coated refined LS compared to others based on analysis, phenol total, microencapsulation efficiency, yield and phenol release and had high solubility. Microcapsules morphology analysis showed that microcapsules had nearly spherical shape, little shriveled, and without agglomeration with low levels of damage. Microcapsules average diameter was 0.24 μm.

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References

Akhilesh, D., Faishal, G., and Kamath, JV., 2012. Comparative Study of Carriers Used In Proniosomes. Review Article. International Journal of Pharmaceutical and Chemical Sciences. (1): 164–173.

Amerie, M., and Maa, Y.F., 2006. Spray drying of Biopharmaceuticals: Stability and Process Considerations. Drying Technol. 24: 763–768.

Anwar, S.H., and Kunz, B., 2011. The Influence of Drying on Thestabilization of Fish Oil Microcapsules: Comparison of Spraygranulation, Spray drying and Freeze Drying. J. Food Eng. 105: 367–378.

Association of Official Analytical Chemists (AOAC), 1990. Official methods of analysis, method 923.03, ash of flour: Direct method (17th ed.). Arlington, VA.

Association of Official Analytical Chemists (AOAC), 2005.Official Analytical Chemist Official Methods on Analysis.18th Ed, Gaithersburg, Maryland.

Budijanto, S., Rokhani, H., Sulusi, P., Setyadjit., Sukarno., and Ita, Z., 2008. Identifikasi dan Uji Keamanan Asap cair Tempurung Kelapa untuk Produk Pangan. Jurnal Pascapanen, 1 (5): 32–40.

Bhullar, G. S., and Bhullar, N. K., 2013. Agricultural Sustainability: Progress and Prospects in Crop Research. Swiss Federal Institute of Technology Zurich, Switzerland. Academic Press is an imprint of Elsevier.

Bratzler, L. J., Spooner,M. E., Weathspoon, J. B., and Maxey J. A., 1969. Smoke Flavours as Related to Phenol, Carbonil, and Acid Content Of Bologna. Journal of Food Science 34: 146–153.

Bylaite´, E., Venskutonis, P. R., and Mapdbieriene, R., 2001. Properties of Caraway (Carumcarvi L.) Essential Oil Encapsulated into Milk Protein-based Matrices. European Food Research and Technology, 212: 661–670.

Caliskan, G., and Dirim, S. N., 2013.The Effects of The Different Drying Conditions and Theamounts of Maltodextrin Addition during Spray drying of Sumac Extract.Food and Bioproducts Processing, 91: 539–548.

Çam, M., İçyer, N. C., and Erdoğan, F., 2013. Pomegranate Peel Phenolics: Microencapsulation, Storage Stability and Potential Ingredient for Functional Food Development. LWT - Food Science and Technology, 55: 117–123.

Cerdeira, M., Martini, S., and Herrera, M.L., 2005. Microencapsulating Properties of Trehalose and of Its Blends with Sucrose and Lactose. Journal of Food Science, 70 (6):401-408.

Chegini, G.R., and Ghobadian, B., 2005. Effect of Spray drying Conditions on Physical Properties of Orange Juice Powder. Drying Technol. 23: 657–668.

Chin, S. T., Sheikh, A. H. N., Quek, S.Y., Yaakob, B. C. M., Russly A. R., and Dzulkifly, M. H., 2010. Effect of Thermal Processing and Storage Condition on TheFlavour Stability of Spray-Dried Durian Powder. Food Science and Technology Journal, (43): 856–861.

Cvitanović, B. A., Stojanović, R., Manojlović, V., Komes, D., Cindrić, I. J., Nedović, V., and Bugarski, B., 2011. Encapsulation of Polyphenolic Antioxidants from Medicinal Plant Extracts in Alginate–Chitosan System Enhanced with Ascorbic Acid by Electrostatic Extrusion. Food Research International, (44): 1094–1101.

Darmadji, P., 1996. AntibakteriAsapCair Dari LimbahPertanian. Agritech 16 (4): 19–22.

Dwiyitno and Rudi, R., 2006. Studi Penggunaan Asap Cair untuk Pengawetan Ikan Kembung (Rastrelligerneglectus) Segar. Jurnal Pascapanen dan Bioteknologi. Kehutanan dan Perikanan 2 (1): 143–148.

Gharsallaoui, A., Roudaut, G., Chambin, O., Voilley, A., and Saurel, R., 2007. Applications of spray-drying in microencapsulation of food ingredients: An overview. Food Research International, 40: 1107–1121.

Girard, J.P., 1992. Smoking in Technology of Meat Products. New York: Clermont Ferrand, Ellis Horwood.

Goula, A.M. and Adamopoulos, K.G. 2004. Influence of Spray drying Conditions on Residue Accumulation–Simulation using CFD. Drying Technology 22: 1107–1128.

Goula, A.M., and Adamopoulos, K.G., 2008. Effect of Maltodextrin Addition during Spray drying of Tomato Pulp in Dehumidified Air II. Powder properties. Drying Technol., 26 (6): 726–737.

Gustavo, V., and Canovas.B., 1999. Food Powders: Physical Properties, Processing, and Functionality. Spinger publisher. Texas.

Hogan, S. A., McNamee, B. F., O'Riordan, E. D., and O'Sullivan, M., 2001. Micro-encapsulation Properties of Sodium Caseinate. Journal of Agricultural and Food Chemistry, 49: 1934–1938.

Honarkar, H. and Barikani, M., 2009. Applications of Biopolymers I: Chitosan. Monatsh Chemistry, 140: 1403–1420 .

Hsieh, Wen-Chuan, Chang, Chih-Pong, and Gao, Ying-Lin, 2006. Controlled Release Properties of Chitosan Encapsulated Volatil Citronella Oil Microcapsules by Thermal Treatments. Colloids and Surfaces B: Biointerfaces, 53: 209–214.

Kažimírová and Jablonická, 1994. Evaluation of Potential Mutagenic Effect of The Liquid Smoke Preparation UTP In Vivo: Cytogenetic Analysis of Mouse Bone Marrow. Mutation Research, 323: 89–92.

Kim, Y.D., Morr, C.V., Schenz, T.W., 1996. Microencapsulation Properties of Gum Arabic and Several Food Proteins: Spray-Dried Orange Oil Emulsion Particles. J. Agric.Food

Chem., 44: 1314–1320.

Krishnaiah, D., Sarbatly, R., and Nithyanandam, R., 2012. Microencapsulation of Morindacitrifolia L. Extract by Spray-Drying. Chemical Engineering Research and Design, 90: 622–632.

Lim, W. J., Liang, Y. T., Seib, Y. T., and RAO, C. S., 1992.Isolation of Oat Starch from Oat Flour. Cereal Chem., 69 (3):233-236.

Loksuwan, J., 2006. Characteristics of Microencapsulated β-Carotene Formed by Spray drying with Modified Tapioca Starch, Native Tapioca Starch and Maltodextrin. Food Hydrocolloids, 21: 928–935.

Maga, J. A., 1998. Smoke in Food Processing. CRC Press. Florida.

Marchal LM, Beeftink HH, Tramper J (1999). Towards A Rational Design of Commercial Maltodextrins. Trends Food Sci. Technol. 10: 345–355.

Nesterenko, A., Alric, I., Silvestre, F., and Durrieu, V., 2013. Vegetable Proteins in Microencapsulation: A Review of Recent Interventions and Their Effectiveness. Review. Industrial Crops and Products, 42: 469–479.

Panagan, A.T and Nirwan, S., 2009. Uji Daya Hambat Asap Cair Hasil Pirolisis Kayu Pelawan (Tristaniaabavata) Terhadap Bakteri Echerichia coli. Jurnal Penelitian Sains Edisi Khusus Desember© 09-12-06.

Popa, M.I., Aelenei, N., Popa, V.I., and Andrei, D., 2000. Study of Interaction Between Polyphenolic Compounds and Chitosan. Reactive and Fuctional Polymers, 45: 35–43.

Pszczola, D. E., 1995. Tour Highlights Production and Uses of Smoke-Based Flavors. Food Technology, 49 (1): 70-74.

Şahin-Nadeem, N., Torun, M., and Özdemir, F., 2011. Spray drying of The Mountain Tea (SideritisStricta) Water Extract by Using Different Hydrocolloid Carriers. LWT - Food Science and Technology, 44: 1626–1635.

Şahin-Nadeem, H., Dinçer, C., Torun, M., Topuz, A., and Özdemir, F., 2013. Influence of Inlet Air Temperature and Carrier Material on The Production of Instant Soluble Sage (Salvia Fruticosa Miller) by Spray drying. LWT - Food Science and Technology, 52: 31–38.

Saloko, S., Purnama, D., Setiaji, B., and Pranoto, Y., 2012.Structural Analysis of Spray-Dried Coconut Shell Liquid Smoke Powder.J.Teknol. danIndustriPangan, XXIII (2): 173–178.

Saloko, S., Purnama, D., Setiaji, B., and Pranoto, Y., 2014.Antioxidative and Antimicrobial Activities of Liquid Smoke Nanocapsules Using Chitosan and Maltodextrin and Its Application on Tuna Fish Preservation. Food Bioscience, 7: 71–79.

Senter, S. D., Robertson, J. A., and Meredith, F. I., 1989. Phenolic Compounds of The Mesocarp of Cresthaven peaches during Storage and Ripening. Journal of Food Science, 5 (54): 1259–1268.

Shaikh, J., Rajesh, B., and Rekha, S., 2006. Microencapsulation of Black Pepper Oleoresin. Food Chemistry, (94): 105–110.

Shi, Y.-C., Eden, J. L., Kasica, J. J., and Jeffcoat, R., 2000. High Solids, Single Phase Process for Preparing Enzyme-Converted Starches.United State Patent No. 6.054.302.

Shu, B., Yu, W., Zhao, Y., and Liu, X., 2006. Study on Microencapsulation of Lycopene by Spray-drying. Journal of food engineering, 76: 664–669.

Storz, E., Steffens, K., 2004. Feasibility Study for Determination of The Dextrose

Equivalent (DE) of Starch Hydrolysis Products with Near-Infrared Spectroscopy (NIRS). Starch 56 (2): 58–62.

Teixeira, M. I., Andrade, L. R., Farina, M., and Rocha-Leao, M. H. M., 2004.Characterization of Short Chain Fatty Acid Microcapsules Produced by Spray drying. Materials Science and Engineering, 24: 653–658.

Thies, Curt, 2001. Microencapsulation.Kirk-Othmer Encyclopedia of Chemical Technology.Vol. 16.John Wiley & Sons, Inc.

Tonon, Renata V., Brabet, C., and Hubinger, Míriam D., 2008. Influence of Process Conditions on The Physicochemical Properties of Açai (Euterpeoleraceae Mart.) Powder Produced by Spray drying. Journal of Food Engineering, 88: 411–418.

Wang, Y-J.and Wang, L., 2000. Structures and Properties of Commercial Maltodextrins from Corn, Potato, and Rice Starches. Starch/Stärke, 52: 296–304.

Yusraini, E., Hariyadi, P., and Kusnandar, F., 2013.Preparation and Partial Characterization of Low Dextrose Equivalent (DE) Maltodextrin from Banana Starch Produced by Enzymatic Hydrolysis. Starch/Stärke, 65: 312–321.