

JURNAL INTEGRASI PROSES

Website: http://jurnal.untirta.ac.id/index.php/jip

Submitted : 24 Februari 2020

Revised : 23 Maret 2020

Accepted : 2 Juni 2020

ENCAPSULATION BIOACTIVE COMPUND PROPOLIS WITH CARRAGEENAN-GUM ARABIC BY SPRAY DRYING

Denni kartika sari^{1*}, Retno sulistyo Dhamar Lestari¹, Prasetyo Khinanta¹, Muhammad Sahlan²

¹Department of Chemical Engineering, Faculty of Engineering, Universitas Sultan Ageng Tirtayasa

Jln Jenderal Sudirman Km 3, Cilegon, Indonesia

²Department of Chemical Engineering, Faculty of Engineering, Universitas Indonesia, Depok

16424, Indonesia

*Email: denni.kartikasari@untirta.ac.id

Abstract

Propolis is a product from the honeybee, particularly from flowers, leaf buds, plant resins that bee collected as a building material. Propolis exerts numerous pharmacological activities such as antioxidant, antibacterial, anticancer, antifungal, anti-inflammatory, antiviral, etc. There is growing interest in the food industry in microencapsulation to protect, isolate, or control the release of given substances. Spray drying is one of many methods to encapsulate food ingredients. Spray-drying technique is used in this study. A solution of k-carrageenan gum arabic and maltodextrin is used as the binding materials. In this work, the effects of spray-drying on the encapsulation yield, particle size, and total phenolic content of the bioactive components of propolis were determined for a different concentration between carrier agent and propolis at volume ratio 1:0,5 1:1, 1:1,5,1. The SEM testing showed that the microcapsules had a regular spherical shape with the size 1 μ m. The spray-dried propolis extract showed high total phenolic content 67,78 mg (GAE)/g extract at ratio 1:2, and the highest yield at concentration ratio between the carrier agent and propolis 1:0,5 is 64,7 %.

Keywords: Carrageenan, Propolis, Spray drying

1. INTRODUCTION

1. There has been increased interest, especially in dried extracts based on natural products. One of them is the propolis product (Manojlović, Nedović, Kailasapathy, & Zuidam, 2010). Propolis is a resin with variable color and consistency. Bees collect it from different parts of plants, such as flower buds and resin exudates. Propolis presents a strong and characteristic smell due to its volatile phenolic acid fraction, strong adhesive properties and complex chemistry (55% of resins and balsams, 30% of wax, 10% of volatile oils and around 5% of pollen), as well as mechanical impurities (Daniel-da-Silva, Ferreira, Gil, & Trindade, 2011). The spray drying process allowed obtaining propolis in the powder form. It would preserve antioxidant activity. Spray drying also increased stability during storage at room temperature had low

hygroscopicity. It was highly dispersible in cold water besides in addition to maintaining the content of antioxidants.

2. The drying process plays an essential role in the preservation of agricultural drying, for better preservation, easier handling, and a reduced bulk volume for transportation (Fudholi et al., 2011). The spray-drying process is used in the food industry to produce dry powders. The preparation of instant food powders needs a further step of agglomeration after spray drying. The spray drying operation is used to get larger particles (from 50–80 µm to250–500 µm). In microencapsulation by spray drying, a target compound is combined with one or more 'wall' materials to form an emulsion or dispersion (Gianfrancesco, Turchiuli, & Dumoulin, 2008). Food ingredients such as vitamins, flavors, starter cultures

carotenoids, fats, and oils could be encapsulated using spray drying techniques (Wilson & Shah, 2007).

The encapsulation could be defined as a process to entrap one substance within another substance. It also is known microencapsulation is a physical process in which the active ingredient (core material). Solid particles, water, or gas droplets are packed in a secondary material (wall). In this study is used kappa carrageenan, maltodextrin, gum arabic as secondary material (Manojlović et al., 2010). It could also be applied to entrap natural compounds like essential oils or vegetable extracts containing antioxidants and polyphenols (Spigno, Garrido, Guidesi, & Elli, 2015). Microencapsulation is a technique for preserving and facilitating the use of sensitive ingredients. In microencapsulation, the core material is surrounded or embedded in a protective layer of differing composition. A liquid is transformed into a powder in the microencapsulation process. Microencapsulation can mask undesirable flavors or odors, control the release rate and location of a compound, and impact the bioavailability of the encapsulated material. Microcapsules can have a variety of structural types multi-core, single core-shell, wall, multi-wall, continuous matrix. Microencapsulation structure depends on the processing method and materials involved in the preparation of the solution before spray drying. However, there is still limited study for encapsulated propolis, with spray drying technique. This study aims to determine the effect of a carrier agent and propolis in spray drying.

2. MATERIAL AND METHODS

2.1 Material

Materials used are Aquades, Ethanol 96% (Sigma Aldrich), The used s Gum Arab, Kappa Carrageenan, Maltodextrin was a typical household (kingmao), and Propolis. k-carrageenan and Propolis from the same production batch was used for all experiments (Marquiafável et al., 2015).

2.2 Preparation and Spray Drying

The coating material maltodextrin and Arabic gum were prepared as described by (Marquiafável et al., 2015) (Da Silva et al., 2013) with modification. The propolis yield was carried out by drying propolis in an oven at 90°C until only the remaining propolis solids and then weighed, a coating matrix comprising 1 g of maltodextrin, 1 g of Carrageenan and 0.2 gr Gum Arabic mixed with 200 ml aquadest using and then Homogenizer using Ultra Turrax. With Stirring, speed is 16000 rpm for 2 minutes. Propolis solution is made by mixing pure propolis with ethanol with a ratio of 1: 4. The propolis solution was mixed on an emulsion of 30 ml, 60 ml, 90 ml, and 120 ml volumes using Homogenizer Ultra Turrax with a stirring speed of 16000 rpm for 2 min. The emulsion then enters the spray dryer. The sample is inserted into a glass beaker. The operating conditions of the equipment are 20% pump pressure, nitrogen gas pressure-temperature 150°C.

2.3 Total Phenolic Content (TPC)

Total phenolics (TPC) of the samples were determined spectrophotometrically by the Folin-Ciocalteu reagent according to the method of (Bobo-García et al., 2015).

2.4 Viscosity

Viscosity was measured using the method as follows (Ajithkumar, 2017): Sample of 7.5 grams and then stirred using a magnetic stirrer. Dispersed with agitation for 10 to 20 min in 450 ml of deionized water. The solution was agitated for 10-15 minutes at room temperature, and heat to a temperature of 80°C measuring viscosity using spindle two at 30 rpm spindle rpm and wait until stabilized (5-6 rounds).

3. RESULTS AND DISCUSSION

The pseudoplastic behavior of kappa-carrageenan can be used as a plasticizer. It made the microencapsulate round and smooth and increase the adhesion force between walls and core materials. Also, kappa carrageenan has other properties as an emulsifier, safe to eat, and biodegradation (Purnomo, Khasanah, & Anandito, 2014). Gum Arabs added in emulsion because of its high solubility. The main characteristic of gum Arab is texture-forming, filmforming, fastener, and emulsifier, which is good with the presence of protein components in gum Arabs. Gum Arabs can retain flavor from dried foods by method spray drying because this gum can form a protective layer of decomposition change (Bénech, 2008). Gum Arabs have weaknesses. The price is quite high and limited availability as well its oxidation resistance is low

The use of gum Arab mixed with dextrin such as maltodextrin. Maltodextrin is a starch hydrolysate product with an average chain length of 5-10 units/glucose molecule. Maltodextrin is composed of glucose units and is not sufficient for stabilizing oil or flavor in solution viscosity. Therefore, maltodextrin is usually combined with ingredients such as gum Arab or other modified starch for stability purposes (Styles, 2003). Gum Arabs in emulsions could produce microcapsules that have high retention, but it had low oxidation resistance. Therefore, the use of gum Arab can be combined with maltodextrins with high oxidation resistance Maltodextrin has good solubility in water and low viscosity values even at high concentrations. These properties make maltodextrin useful for the coating material. Maltodextrins are deficient in terms of emulsification property and surface-active features (Akdeniz, Sumnu, & Sahin, 2017). The addition of the carrier agent in the microencapsulation process could increase total propolis solids. The total content of solids affects the duration of the drying process and the resulting yield.

Microencapsulation combined with kappa carrageenan. It can increase the yield of microencapsulation due to the high molecular weight of carrageenan that is above 100 kDa or ranged between 100-800 thousand kDa (DeMan, 1999). The nature of kappa-carrageenan is the fraction capable of forming a gel in water and increasing the viscosity of the solution so that the total dissolved solids become increased, which results in a higher yield compared to the ratio of other coating combinations, therefore, required viscosity test. Besides, viscosity testing of the emulsion is essential because the spray dryer used as a specification of emulsion viscosity should be carried 122 cp.

Tabel 1. Emulsion viscosity		
Volume propolis in total volume of emulsion (ml)	Viscosity of emulsion (cp)	
30	31.40	
60	29.20	
90	27.30	
120	20.50	

The excessively high viscosity of the solution can damage the nozzle and complicate the atomizer's process so that it can become unstable in the flow within the spray drying, resulting in many microencapsulated powders attached to the chamber spray dryer tube. The yield of the spray drying process, which is defined as the ratio of powder mass collected to the mass of total solids in the feed, is an essential indicator of industrial success. Residue formation during spray drying indicates poor process performance and is mainly caused by the stickiness of the product. A yield value higher than 50% as a successful drying process (Guzel-Seydim, Kok-Tas, Greene, & Seydim, 2011). Lower ratio propolis will increased viscosity; higher viscosity will increase the yield of microcapsules due to higher carrier agent concentration (Purnomo et al., 2014). Yield increased along with the addition of coating materials with various ratios combination. From Figure 2, we could have seen a sample with ratio propolis and carrier agent 2:1 much more agglomerated and also supported with figure 3 Scanning electron of particles. The yield of spray drying less than 50% caused stickiness to the product from Tabel 1. It could be seen the yield of less than 50%..

Tabel 2. Yield result			
Emulsion ratio		_	
Propolis	Carrier	Mass (gr)	yield(%)
	agent		
0,5	1	2,1353	64,71
1	1	2,8136	63,95
1,5	1	2,7592	50,17
2	1	2,9970	45,41

From Figure 1. the addition of propolis concentration will increase the total phenolic content because propolis contains considerable amounts of phenolic compounds (Yang et al., 2011). A list of the major classes of chemicals occurring in propolis are resins composed of flavonoids and phenolic acids or their esters, which often form up to 50% of all

ingredients, the total phenolic content also depends the amount of propolis added (Huang, Zhang, Wang, Li, & Hu, 2014).

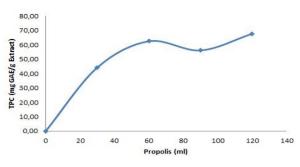


Figure 1. The total phenolic content



Figure 2. Sample of spray-dried propolis from left to right, 0,5:1, 1:1, 1,5:1, 1:2

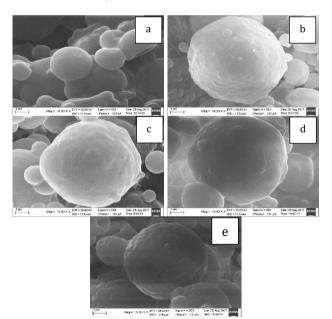


Figure 3. SEM of particles of spray-dried propolis a.(0,5:1), b. (1:1), c.(1,5:1), d.(2:1), e. without propolis

From Figure 3, the sample tended to agglomerate, especially at a volume ratio of 2:1. When the propolis at higher concentration, agglomeration, formed, caused a reversible consolidation of the bridges because groups of particles with structural integrity are formed. The next phase of caking is associated with the loss of integrity. Caking is a result of the enlargement of bridges among the particles, leading to a decrease of the space between particles and group defamation. In the final phase of caking, the sample is liquefied. Storage needs to consider humidity (da Silva, Favaro-Trindade, de Alencar, Thomazini, & Balieiro, 2011). In figure 2, increased propolis content the color of the product getting darker and the moisture content getting higher too.

4. CONCLUSION

It is possible to dried microencapsulation of the propolis by spray drying. In this study, ratio carrier agent and propolis have significant effects at yield and total phenolic content. The highest total phenolic compound is at 67,78 mg (GAE)/g at ratio 1:2, and the highest yield at 1:0,5, which is 64,7%.

5. REFERENCES

- Akdeniz, B., Sumnu, G., & Sahin, S. (2017). The effects of maltodextrin and gum Arabic on encapsulation of onion skin phenolic compounds. Chemical Engineering Transactions. Retrieved from https://doi.org/10.3303/CET1757316
- Bénech, A. (2008). Gum Arabic A functional hydrocolloid for beverages. Agro Food Industry Hi-Tech.
- Bobo-García, G., Davidov-Pardo, G., Arroqui, C., Vírseda, P., Marín-Arroyo, M. R., & Navarro, M. (2015). Intralaboratory validation of microplate methods for total phenolic content and antioxidant activity on polyphenolic extracts, and comparison with conventional spectrophotometric methods. Journal of the Science of Food and Agriculture. Retrieved from https://doi.org/10.1002/jsfa.6706
- Da Silva, F. C., Da Fonseca, C. R., De Alencar, S. M., Thomazini, M., Balieiro, J. C. D. C., Pittia, P., & Favaro-Trindade, C. S. (2013). Assessment of production efficiency, physicochemical properties and storage stability of spray-dried propolis, a natural food additive, using gum Arabic and OSA starch-based carrier systems. Food and Bioproducts Processing. Retrieved from
- https://doi.org/10.1016/j.fbp.2012.08.006 da Silva, F. C., Favaro-Trindade, C. S., de Alencar, S. M., Thomazini, M., & Balieiro, J. C. C. (2011). Physicochemical properties, antioxidant activity
- and stability of spray-dried propolis. Journal of ApiProduct and ApiMedical Science. Retrieved from https://doi.org/10.3896/ibra.4.03.2.05
- Daniel-da-Silva, A. L., Ferreira, L., Gil, A. M., & Trindade, T. (2011). Synthesis and swelling behavior of temperature responsive κ-carrageenan nanogels. Journal of Colloid and Interface Science. Retrieved from https://doi.org/10.1016/j.jcis.2010.12.071
- DeMan, J. M. (1999). Principles of Food Chemistry. Science.
- Fudholi, A., Othman, M. Y., Ruslan, M. H., Yahya, M., Zaharim, A., & Sopian, K. (2011). The effects of drying air temperature and Humidity on the drying kinetics of seaweed. In Recent Researches in Geography, Geology, Energy, Environment and

Biomedicine - Proc. of the 4th WSEAS Int. Conf. on EMESEG'11, 2nd Int. Conf. on WORLD-GEO'11, 5th Int. Conf. on EDEB'11.

- Gianfrancesco, A., Turchiuli, C., & Dumoulin, E. (2008). Powder agglomeration during the spray-drying process: Measurements of air properties. Dairy Science and Technology. Retrieved from https://doi.org/10.1051/dst:2007008
- Guzel-Seydim, Z. B., Kok-Tas, T., Greene, A. K., & Seydim, A. C. (2011). Review: Functional properties of kefir. Critical Reviews in Food Science and Nutrition. Retrieved from https://doi.org/10.1080/10408390903579029
- Huang, S., Zhang, C. P., Wang, K., Li, G. Q., & Hu, F. L. (2014). Recent advances in the chemical composition of propolis. Molecules. Retrieved from https://doi.org/10.3390/molecules191219610
- Manojlović, V., Nedović, V. A., Kailasapathy, K., & Zuidam, N. J. (2010). Encapsulation of probiotics for use in food products. In Encapsulation Technologies for Active Food Ingredients and Food Processing. Retrieved from https://doi.org/10.1007/978-1-4419-1008-0_10
- Marquiafável, F. S., Nascimento, A. P., Barud, H. da S., Marquele-Oliveira, F., de-Freitas, L. A. P., Bastos, J. K., & Berretta, A. A. (2015). Development and characterization of a novel standardized propolis dry extract obtained by factorial design with high artepillin C content. Journal of Pharmaceutical Technology and Drug Research. Retrieved from https://doi.org/10.7243/2050-120x-4-1
- Purnomo, W., Khasanah, L. U., & Anandito, B. K. (2014).
 Pengaruh Ratio Kombinasi Maltodekstrin, Karagenan dan Whey Terhadap Karakteristik Mikroenkapsulan Pewarna Alami Daun Jati (Tectona Grandis L. F.). Jurnal Aplikasi Teknologi Pangan.
- Ajithkumar, S. (2017). Optimization Of Carrageenan Extraction Process From Seaweed. World Journal of Pharmacy and Pharmaceutical Sciences. Retrieved from https://doi.org/10.20959/wjpps20174-9034
- Spigno, G., Garrido, G. D., Guidesi, E., & Elli, M. (2015). Spray-drying encapsulation of probiotics for icecream application. Chemical Engineering Transactions. Retrieved from https://doi.org/10.3303/CET1543009
- Styles, B. T. (2003). Encyclopedia of Food Sciences and Nutrition. Encyclopedia of Food Sciences and Nutrition. Retrieved from https://doi.org/10.1016/B0-12-227055-X/00927-5
- Wilson, N., & Shah, N. P. (2007). Microencapsulation of vitamins. International Food Research Journal.
- Yang, H., Dong, Y., Du, H., Shi, H., Peng, Y., & Li, X. (2011). Antioxidant compounds from propolis collected in Anhui, China. Molecules. Retrieved from https://doi.org/10.3390/molecules16043444