Performance of Alginate-Based Coatings in Liquid Smoke Encapsulation

Iffah Muflihati[1*], *Ayuni* Rizma Maulida[1], *Umar* Hafidz Asy'ari Hasbullah[1], *Arief* Rakhman Affandi[1], *Silvia* Dwi Suryani[1] and *Abdillah* Fathan Generus Annajah[1]

[1]Departement of Food Technology, Faculty of Engineering and Informatics, Universitas PGRI Semarang, Semarang, Indonesia

Abstract. Liquid smoke contains bioactive compounds such as phenols, carbonyls, and organic acids that function as antimicrobial and antioxidant agents. However, its major limitation is its high volatility, which requires protection through encapsulation. This study aimed to determine the characteristics of encapsulated liquid smoke using the coacervation technique with various coating materials, including alginate (A), alginate+gum arabic (AA), alginate+chitosan (AK), alginate+tapioca (AT), and alginate+maltodextrin (AM), as well as two drying methods, cabinet dryer (CB) and open air drying (OA). The results showed that the two factors coating material and drying method significantly affected the total phenol, total acidity, total carbonyl content, encapsulation efficiency, and loading capacity. Total phenol range from 0.75–3.20%, total acidity from 1.72–4.33%, and total carbonyl content from 6.26–11.53%. AK combination produced the highest retention of phenols, acids, and carbonyls, as well as the highest encapsulation efficiency and loading capacity.

1 Introduction

Liquid smoke results from a pyrolysis process obtained from vapor condensation. Liquid smoke contains compounds that can be used as antimicrobials, antifungals, and flavor enhancers. One of the compounds in liquid smoke that acts as an antibacterial agent is antioxidants such as aldehydes, carboxylic acids, and phenols. Although the use of liquid smoke in the food industry in Indonesia remains limited, liquid smoke has great potential as a natural food additive. This is due to the antioxidant compounds in liquid smoke and its low health risks. Therefore, liquid smoke can be used as an alternative in food additives such as antimicrobial agents and flavorings supported by specific aroma attributes such as sweet, floral, fruity, spicy, broth, and meat. The active compounds in liquid smoke are susceptible to evaporation, making storage and distribution less efficient. The phenolic content in liquid smoke can be damaged by oxidation. Encapsulation is a method that can be used to minimize the weaknesses of liquid smoke. This method involves coating the core material (liquid smoke) with a coating material for the purposes of protection, isolation, and facilitating storage and controlled release. Encapsulation can also be useful for reducing degradation and

---

[*] Corresponding author: iffahmuflihati@upgris.ac.id

aroma loss during processing, storage, and summarizing volatile components before application in food products.

The liquid smoke encapsulation process usually uses a spray dryer, but this process is costly, so coacervation is used as an alternative. The advantages of the coacervation technique are that the process is easy, the equipment is simple, it can be done at room temperature, and it has a high efficiency value. The encapsulation technique is closely related to the coating material used to wrap the core material and trap the core material. The coating material will affect the success of the encapsulation process, which is influenced by the properties of the coating material, the stability of the core material, and the suitability of the encapsulation method. Therefore, alginate is used as a coating material because it has biodegradability, biocompatibility, and is capable of forming chain associations. The presence of divalent cations in alginate can form a third-dimensional gel. However, alginate contains different monovalent ions, which cause alginate to perform less than optimally and require the addition of other coating materials. The addition of tapioca as a coating material can support the structure that can modulate bead shrinkage and surface texture after drying. Besides tapioca, maltodextrin is also used as a coating material because it is heat-resistant, low-cost, has a neutral aroma and taste, low viscosity at high solids, and can retain antioxidant content. The combination of alginate with tapioca and maltodextrin is expected to support better encapsulation and retain the compounds contained in the liquid smoke.

The type of drying also affects the encapsulation technique in drying the encapsulant. In this study, two types of drying were used, cabinet drying (CB) and open air drying (OA). Cabinet dryer drying has the advantage of controllable drying time and temperature, which affects the quality of the final product. Meanwhile, drying using air drying has the advantage of minimizing damage to materials due to the heat generated from the drying process. This study aims to determine the characteristics of liquid smoke encapsulation using the coacervation technique combined with different coating materials and drying methods.

2 Methods

Research using a factorial design method with two factors, the coating material consisting of alginate (A), alginate + gum arabic (AA), alginate + chitosan (AK), alginate + tapioca (AT), and alginate + maltodextrin (AM) with a ratio of 1:1, amounting to 1.5 grams, except for alginate, which is 3 grams. The second factor was the type of drying, consisting of cabinet dryer (CB) and open air drying (OA).

2.1 Production of Liquid Smoke Encapsulation

Aquades 100 mL was heated to 50°C with continuous stirring at a speed of 500 rpm. 1 gram of casein was homogenized for 5 minutes. The coating material was added at a ratio of 1:1 and homogenized for 5 minutes at a temperature of ±75°C. The homogenized suspension was rested at room temperature for 30 minutes. The suspension that has been stirred continuously is then added with 1 mL of liquid smoke and agitated until homogeneous for 5 minutes. The liquid smoke encapsulation suspension is placed in OPP plastic. Alginate (A) was used to encapsulate liquid smoke to result hydrogel beads. The beads were placed on baking sheet lined with aluminium foil followed by two drying methods: i) a cabinet drying (CB) at 50C, RH 20% for 6h, and ii) open air drying (OA) at 30C; RH 75% for 24h. Samples were collected and stored in a plastic container until analysis.

2.2 Phenol Content

0.4 grams of microcapsules diluted in 10 mL of distilled water, 1 mL taken and diluted in 10 mL. The dilution result was measured using a micropipette and 1 mL samples were placed in a test tube. 1 mL of Na_2CO_3 was added and stored at room temperature. 1 mL of Folin-Ciocalteu reagent was homogenized in 7.5 mL of distilled water using a vortex and kept at room temperature for 30 minutes. The sample is measured at a wavelength of 770 nm. The phenol level of the sample is calculated based on the standard curve equation for gallic acid using the formula:

$$\text{Phenol Total (\%)} = (x.1000.\text{sample volume}.100)/(1000.\text{sample weight})$$

Notes :
x in the equation y = ax+b in the standard curve

2.3 Total Acidity

Total acid calculation using the titration method. 1 mL of liquid smoke is mixed with 100 mL of distilled water until homogeneous. 1 mL is diluted to 100 mL. 3 droplets of PP are introduced into the sample, then titrated with 0.1 N NaOH until a pink color is obtained. Total acid calculation using the formula:

$$\text{Total Acidity (\%)} = ((\text{mL NaOH} \times \text{N NaoH} \times \text{BM acetic acid} \times fp)/\text{ mg sample}]\times 100\%$$

Notes :
N NaoH : Normality of NaOH
BM : Molar mass of acetic acid
Fp : Dilution Factor

2.4 Carbonyl Content

1 mL of liquid smoke is diluted in 25 mL of distilled water. 1 mL of sample is sampled and diluted to 100 mL. 1 mL of the diluted sample is mixed with 1 mL of 2,4-dinitrophenylhydrazine and 1 drop of concentrated HCl, heated to 50°C for 30 minutes. After the mixture cools, 8 mL of 1 N KOH solution and measure at a wavelength of 480 nm. Preparation of standard curve 1.265 mL of pure acetone is diluted to 100 mL, 1 mL is taken and diluted to 100 mL. The 10 mL is then diluted to 20 mL (the final concentration is 50 ppm, called solution A). Each solution was added with 1 mL of 2,4-dinitrophenylhydrazine and 1 drops of concentrated HCl, heated for 30 minutes at 50°C, the mixture was cooled and added with 8 mL of 1 N KOH solution, then absorbance was measured at a wavelength of 480 nm. Calculation of carbonyl content using the equation :

$$\text{Carbonyl Content} = [(X \times fp)/\text{mg sample}] \times 100\%$$

2.5 Encapsulation Efficiency

Encapsulation efficiency can be calculated based on total phenol. Encapsulation efficiency is calculated as the ratio between the phenol content in the liquid smoke microcapsules and the phenol content in the liquid smoke using a specific formula. Encapsulation efficiency is calculated using the equation :

$$\text{Encapsulation Efficiency (\%)} = (\text{microcapsule phenol}/\text{initial phenol}) \times 100\%$$

2.6 Loading Capacity

Loading capacity (L) is the weight of phenol in liquid smoke microcapsules (KT) per weight of microcapsules (W) [1]. Loading capacity can be calculated by dividing KT by W.

$$\text{Loading Capacity (\%)} = (\text{weight of phenol in liquid smoke microcapsules/ weight of microcapsules}) \times 100\%$$

$$KT = \%\ \text{Phenol} \times \text{weight of microcapsules}$$

2.7 Data Analysis

Data analysis was performed using the Independent-Samples T-Test. If the results showed a difference, the DMRT test was performed with a 95% confidence level. Data analysis was conducted using SPSS version 21.

3 Result and discussion

The results of this study include total phenols, total acids, total carbonyls, encapsulation efficiency, and loading capacity in encapsulated liquid smoke. The analysis results are presented in Table 1 and Table 2.

Table 1. Characteristics of Encapsulated Liquid Smoke

Coating Materials	Phenol Content (%)		Total Acidity (%)		Carbonyls Content (%)	
	CB	OA	CB	OA	CB	OA
A	$2,23 \pm 0,07^{cA}$	$0,87 \pm 0,05^{bB}$	$2,24 \pm 0,28^{aA}$	$2,30 \pm 0,31^{bcA}$	$10,47 \pm 0,34^{cA}$	$6,42 \pm 0,42^{abB}$
AA	$1,65 \pm 0,08^{bA}$	$0,75 \pm 0,03^{aB}$	$2,27 \pm 0,30^{abA}$	$2,80 \pm 0,30^{cA}$	$7,66 \pm 0,43^{bA}$	$7,70 \pm 0,19^{cA}$
AK	$3,20 \pm 0,09^{dA}$	$2,05 \pm 0,03^{cB}$	$4,33 \pm 0,30^{cA}$	$2,24 \pm 0,00^{bB}$	$8,15 \pm 0,14^{bA}$	$7,11 \pm 0,27^{bcB}$
AT	$1,36 \pm 0,01^{aA}$	$0,81 \pm 0,01^{abB}$	$2,79 \pm 0,29^{bA}$	$2,44 \pm 0,30^{bcA}$	$6,26 \pm 0,41^{aA}$	$6,42 \pm 0,47^{abA}$
AM	$1,28 \pm 0,12^{aA}$	$0,75 \pm 0,10^{aB}$	$2,82 \pm 0,31^{bA}$	$1,72 \pm 0,30^{aB}$	$11,53 \pm 0,33^{dA}$	$6,31 \pm 0,41^{aB}$

Notes : Alginate (A), Alginate+Gum Arabic (AA), Alginate+Chitosan (AK), Alginate+Tapioca (AK), Alginate+Maltodextrin (AM), Cabinet Dryer (CB), and Open Air Drying (OA). Lowercase letters in the same column for each analysis indicate results that are significantly different ($P<0.05$) as tested using One Way Anova. Uppercase letters in the same column for each analysis indicate results that are significantly different ($P<0.05$) as tested using Independent-Samples T Test.

3.1 Phenol Content

According to Table 1, the phenol content of encapsulated liquid smoke varies between 0.75 and 3.20%, which indicates a significant difference between alginate and other types of coating materials. The total encapsulated liquid smoke phenol in this study was higher than in studies [2][3], due to the bonds formed by alginate and chitosane, protecting the phenol compounds in the capsules. The combination of AK coating material has an effect on the formation of polyelectrolyte complexes that have the ability to encapsulate more phenolic compounds compared to other coating materials. AA coating material has the lowest total phenol value with air drying due to the lack of bonds between alginate and gum arabic. These factors cause the coating material to be ineffective in retaining phenolic compounds in the capsules. Drying CB with AM coating material is ineffective in retaining phenolic compounds in the capsules due to the reaction of phenolic compounds, which have the ability to enter the polysaccharide structure through the polymer membrane, thereby reducing the ability to retain phenolic compounds in the capsules [4]. The use of high temperatures also affects the microencapsulation process, while low temperatures can cause oxide reactions to proceed more slowly [2]. The phenolic compounds in the sample can be destroyed if drying is insufficient before the formation of particles when low temperatures are applied. The coating material that was used improved the encapsulation efficiency at a phenol level indicating that the effect was more dominant resulting in the highest value. The drying method also contributed to the stability of phenol against oxidation during the drying process.

3.2 Total Acidity

The total encapsulated liquid smoke acid contained ranges from 1.72-4.33%, showing a significant difference between coating materials. The presence of acid contained in liquid smoke can affect the overall organoleptic quality and function as a preservative. Liquid smoke contains acid group compounds and their derivatives, such as alcohol, aldehyde, hydrocarbon, ketone, phenol, and pyridine, which are important to identify the acid content in liquid smoke encapsulation [5]. Coating materials such as AK are important because of the interaction between acetic acid and chitosan through hydrogen bonds, thus increasing the total acid contained. AA is able to absorb acidic compounds due to the reaction of glutamic acid (C=O) bonds with gum arabic, resulting in 2 COOH groups and the formation of NH_2 [6]. The acids contained in liquid smoke are volatile, therefore the type of drying affects the total acid content of the product. High temperatures cause the acidic compounds to evaporate, resulting in low total acid values. This is also due to the duration of heat application; the longer the heat is applied, the more acidic compounds evaporate. Total acid content is affected by differences in coating materials, as these can bind and retain acidic compounds in the material. Meanwhile, the drying process has an impact on acid loss during drying due to differences in temperature and drying time.

3.3 Carbonyl Content

Total carbonyl content is shown in Table 1. it varies between 6.26 and 11.53%, showing that there is a significant difference between coating material A and other coating materials. The highest total carbonyl content in CB drying and coating material AM is due to strong bonds that maintain total carbonyl content during the drying process. Maltodextrin has high water solubility and low viscosity, which can contribute to maintaining the total carbonyl in the encapsulated product [7]. The addition of alginate coats the liquid smoke, resulting in a high total carbonyl. This differs from the OA drying method due to differences in the ambient temperature during the drying process. Higher temperatures tend to reduce volatile compounds in the material. The carbonyl compounds contained in liquid smoke can survive at high temperatures due to the presence of polar groups [7]. Polar groups are higher boiling points than other hydrocarbon molecules, thereby maintaining carbonyl compounds [5].

Table 2. Characteristics of Encapsulated Liquid Smoke

Coating Materials	**Encapsulation Efficiency (%)**		**Loading Capacity (%)**	
	CB	**OA**	**CB**	**OA**
A	32,20±4,11^{bA}	14,04±1,41^{aB}	2,23±0,07^{cA}	0,87±0,05^{bB}
AA	58,56±1,34^{dA}	18,31±2,30^{aB}	1,65±0,08^{bA}	0,75±0,04^{aB}
AK	81,77±2,92^{eA}	34,68±5,23^{cB}	3,20±0,09^{dA}	2,05±0,03^{cB}
AT	19,84±1,30^{aA}	13,00±1,28^{aB}	1,36±0,01^{aA}	0,81±0,01^{abB}
AM	42,97±4,36^{cA}	25,51±2,72^{bB}	1,28±0,12^{aA}	0,75±0,11^{aB}

Notes : Alginate (A), Alginate+Gum Arabic (AA), Alginate+Chitosan (AK), Alginate+Tapioca (AK), Alginate+Maltodextrin (AM), Cabinet Dryer (CB), and Open Air Drying (OA). Lowercase letters in the same column for each analysis indicate results that are significantly different ($P<0.05$) as tested using One Way Anova. Uppercase letters in the same column for each analysis indicate results that are significantly different ($P<0.05$) as tested using Independent-Samples T Test.

3.4 Encapsulation Efficiency

Based on Table 2, the efficiency of encapsulated liquid smoke ranged from 13.00 to 81.77%, showing a significant difference. AK with CB and OA drying had the highest values due to the stronger bond formed between alginate and chitosan. This is in accordance with the highest total phenol results in AK coating material. Encapsulation efficiency is determined by polyphenol compounds the more total phenols produced, the higher the encapsulation efficiency [8]. However, this was not the condition for AT coating material because the interaction between coating materials caused encapsulation efficiency to decrease. Furthermore, the ratio employed also affects the coating material's ability to retain the core material's compound content during the encapsulation process, resulting in low encapsulation efficiency [9]. Encapulation efficiency results indicate that coating materials are a more dominant factor than drying methods. Coating materials are capable of interacting with active compounds, thereby determining the ability to encapsulate compounds. Drying methods support efficiency by reducing moisture content, increasing encapsulation stability, and increasing the concentration of bioactive compounds during the drying process [10].

3.5 Loading Capacity

The loading capacity values produced by encapsulated liquid smoke ranged from 0.75 to 3.20%, which showed a significant difference between coating materials and drying methods. Loading capacity is highly correlated with total phenol; the higher the total phenol produced, the higher the loading capacity value. Therefore, AK had the highest loading capacity value. The main material to be encapsulated with polyphenol compound content also affects the loading capacity [11]. The drying method affects the loading capacity value produced because heating tends to remove volatile components during the encapsulation process [12]. The CB drying method in this study was able to maintain the phenol content in the liquid smoke encapsulation, resulting in a higher loading capacity value compared to the air drying method

4 Conclusion

This research shows that the characteristics of encapsulated liquid smoke are affected by two factors, which are the coating material and the type of drying, affecting total phenols, total acids, total carbonyls, encapsulation efficiency, and loading capacity. The combination of alginate and chitosan coating materials produced the best results due to strong polymer interactions that were able to retain the volatile components. The drying method using a cabinet dryer is effective in preserving active compounds compared to open air drying. On the whole, the combined use of the appropriate coating material and drying method can improve the stability and quality of the chemical components of the encapsulated liquid smoke. Furthermore, the characteristics of the encapsulation produced through the application of the encapsulation product in other food products ought to be further improved. Therefore, droplet measurement during the processing stage ought to be more precise.

References

1. Z. Yang et al., Development and evaluation of novel flavour microcapsules containing vanilla oil using complex coacervation approach. Food Chem **145**, 272–277 (2014). https://doi.org/10.1016/j.foodchem.2013.08.074
2. D. I. Ariestya, F. Swastawati, and E. Susanto, Antimicrobial Activity of Microencapsulation Liquid Smoke on Tilapia [Oreochromis Niloticus (Linnaeus, 1758)] Meat for Preservatives in Cold Storage (± 5 C°). Aquat Procedia **7**, 19–27 (2016). https://doi.org/10.1016/j.aqpro.2016.07.003
3. S. Saloko, P. Darmadji, B. Setiaji, Y. Pranoto, and A. K. Anal, Encapsulation of coconut shell liquid smoke in chitosan-maltodextrin based nanoparticles. Int Food Res J **20**, 3, 1269–1276 (2013).
4. H. Novianty, P. Darmadji, and Y. Pranoto, Utilization of Alginate As an Encapsulation. Mar Res Indones **40**, 1, 1–8 (2015).
5. Budaraga IK, Arnim, Marlida Yetti, and B. Usman, Characteristics of Cinnamon Liquid Smoke Produced Using Several Purification Techniques. Am J Food Sci Nutr Res **3**, 2, 16–21 (2016).
6. Susianti, U. Amalia, and L. Rianingsih, Penambahan Gum Arab dengan Konsentrasi yang Berbeda terhadap Kandungan Senyawa Volatil Bubuk Rusp Ikan Teri (Stolephorus sp.). J Ilmu dan Teknol Perikan **2**, 1, 10–19 (2020). https://doi.org/10.14710/jitpi.2020.8083
7. B. Akdeniz, G. Sumnu, and S. Sahin, The effects of maltodextrin and gum Arabic on encapsulation of onion skin phenolic compounds. Chem Eng Trans **57**, 1891–1896 (2017). https://doi.org/10.3303/CET1757316
8. J. Jayanudin and R. Rochmadi, Pengaruh Perbedaan Bahan Penyalut Terhadap Efisiensi Enkapsulasi Oleoresin Jahe Merah. ALCHEMY J Penelit Kim **13**, 2 (2017). https://doi.org/10.20961/alchemy.v13i2.5406
9. G. Lozano-Vazquez, C. Lobato-Calleros, H. Escalona-Buendia, G. Chavez, J. Alvarez-Ramirez, and E. J. Vernon-Carter, Effect of the weight ratio of alginate-modified tapioca starch on the physicochemical properties and release kinetics of chlorogenic acid containing beads. Food Hydrocoll **48**, June, 301–311 (2015). https://doi.org/10.1016/j.foodhyd.2015.02.032
10. N. Eghbal and R. Choudhary, Complex coacervation: Encapsulation and controlled release of active agents in food systems. LWT - Food Sci Technol **90**, 254–264 (2018). https://doi.org/10.1016/j.lwt.2017.12.036
11. A. Shetta, J. Kegere, and W. Mamdouh, Comparative study of encapsulated peppermint and green tea essential oils in chitosan nanoparticles: Encapsulation, thermal stability, in-vitro release, antioxidant and antibacterial activities. Int J Biol Macromol **126**, 731–742 (2019). https://doi.org/10.1016/j.ijbiomac.2018.12.161
12. N. Kanha, J. M. Regenstein, S. Surawang, P. Pitchakarn, and T. Laokuldilok, Properties and kinetics of the in vitro release of anthocyanin-rich microcapsules produced through spray and freeze-drying complex coacervated double emulsions. Food Chem. **340**, April 2020, 127950 (2021). https://doi.org/10.1016/j.foodchem.2020.127950