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Microencapsulation of Garlic Oil with Gelatin and Maltodextrin Encapsulant Using the Coacervation Method

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ABSTRACT. Garlic Oil is widely used as a raw material for fragrances with volatile active compounds at room temperature. It is easily affected by environmental changes and this problem can be solved by microencapsulation using coacervation methods to protect active compounds. Garlic Oil was encapsulated using gelatin and sodium alginate as coating material with glutaraldehyde as the crosslinking agent. The purpose of this research was to study the effect of the composition of polymers and the mass of garlic oil on encapsulation characteristics. The resulting microcapsules were analyzed using a digital microscope, SEM, FTIR, and encapsulation efficiency. The result showed that microcapsules have an irregular shape with a textured surface. The FTIR spectrum showed an indication of garlic oil with allicin content in microcapsules. The encapsulation efficiency is 34.23% with a yield of 29.43%.

1. INTRODUCTION

The role of plants as a medicinal ingredient is as essential as their role as a foodstuff. One of the plants which can be potentially used as a medicinal ingredient is garlic. *Allium sativum L.* (garlic) is an annual plant that belongs to the Alloidae subfamily from the Amaryllidaceae family [1] Garlic has properties to cure various diseases.

Garlic is one of the plants that can produce essential oil. As it is known that essential oil is a rich and multipurpose oil, the essential oil can act as a stress and anxiety reliever, a treat for insomnia, a remedy for inflammation, and anti micro bacterial agent. Although widely used in various fields, essential oils are susceptible to high temperatures, oxidation, UV rays, and humidity [2]. One of the risks is oxidative damage which may cause off-flavor, shelf-life instability, and also affect sensory properties so solutions are needed to overcome those risks.

After processing, such as cutting, crushing, chewing, or dehydration, the compounds in whole garlic are converted into hundreds of organosulfur compounds in a short period of time, one of which is allicin. Allicin is very sensitive to high temperatures and pH, so encapsulation can be used to increase stability and suppress unpleasant odors [3]. Allicin and other thiosulfinates directly decompose into other compounds, such as diallyl sulfide (DAS), diallyl disulfide (DADS) and diallyl trisulfide (DAT), dithiin and ajoene [4].

Microencapsulation is one of the techniques to protect essential oils from damage during processing and helps stabilize volatile losses. Microencapsulation is a thin coating of solid particles or liquid droplets and liquid dispersion using polymers with particle sizes ranging from 1-5000 μm. Microencapsulation techniques have been widely used to protect food components from a decrease in the content of bioactive compounds, loss of volatile compounds, or to prevent unwanted interactions between ingredients [5]. The coating material can protect the core material, such as oil, which is originally in a liquid form to become a solid so that it is easy to handle and can protect the core material from losing its flavor. One type of essential oil is garlic oil. In its utilization, this oil has not been used to its full potential. Therefore, microencapsulation is required to increase the economic value of garlic oil.

Julaeha, et al., made microcapsules of lime essential oil by complex coacervation with an alginate-gelatin coating on cotton cloth with a citric acid binder to obtain a yield of 46.28%, homogeneous particle size distribution in the range of 1.604 μm, 61.89% oil content, 87.37% encapsulation efficiency, with a smooth surface spherical morphology [6]. Wijesirigunawardana & Perera (2018), made microcapsules of lime peel essential oil with chitosan and Arabian gum coating, which were inserted into the cotton cloth with succinic acid binder and tested on *S. aureus*, *E. coli*, *Bacillus cereus*, and *Salmonella typhimurium* bacteria. The synthesized microcapsules were irregular in shape and differed in size from 15–160 µm with a loading efficiency of 82 ± 4% [7]. Adamiec, et al (2012), made microcapsules of kaffir lime essential oil with konjac glucomannan and Arabian gum coatings, which were tested on the *S. aureus*, *Salmonella Typhimurium*, *E. coli*, *Vibrio cholera*, and *Pseudomonas fluorescens* bacteria and obtained results that microcapsule produced at an inlet temperature of 180°C has the best functional properties [8]. Sharkawy et al (2017), made microcapsules containing limonene and vanillin with chitosan and Arabic gum coatings inserted into the cotton cloth and tested them on *S. aureus* and *E. coli* bacteria. The average diameter of the resulting microcapsules ranged between 10.4 and 39.0 µm, while the resulting encapsulation efficiency was between 90.4% and 100% [9]. Kumari et al., made fragrant textiles containing microcapsules of lime essential oil using acacia gum and gelatin coatings. The microencapsulation results were proven to help control the rate of aroma release and can provide a lasting aroma finish on textiles [10].

Based on those studies, microencapsulation of garlic essential oil with gelatin and maltodextrin coating materials using the coacervation method has never been carried out. The coacervation method was chosen because it is easier and has high encapsulation efficiency. Therefore, the method of making microcapsules with gelatin and maltodextrin can be developed for commercialization so that it can increase the economic value of garlic, gelatin, and maltodextrin. Gelatin was chosen because it has high emulsifying properties which can minimize product size and also can bind oil in water. Maltodextrin was chosen because it has low hygroscopic properties, high solubility in cold water, provides antioxidant properties, and protects volatile materials. The objective of this garlic microencapsulation study was to determine the effect of gelatin and maltodextrin as encapsulants

2. MATERIALS AND METHODS

2.1 Materials

The materials used in the microencapsulation process include garlic oil (Lansida), gelatin (Gelita), maltodextrin (Lihua starch), distilled water (CV. Agung Jaya), tween 20 (Merck), glutaraldehyde (Merck), sodium alginate (CV. Subur Kimia Jaya), acetic acid 98% (Merck), and n-hexane (CV. Agung Jaya).

2.2 Equipment

The equipment used in the microencapsulation process are statives, clamps, thermometers, beakers (Iwaki), magnetic stirrers (Cimarec), pipettes, one set of soxhlet extractors (Iwaki), and Erlenmeyer flask (Iwaki). The equipment used for the characterization process is FTIR (Shimadzu Q-ATR) and SEM (Jeol JCM-7000 model.

2.3 Microencapsulation Process

The microencapsulation process starts with the preparation of polymer solutions. Polymers or polymer mixtures, in this case, gelatin and maltodextrin were varied with a total weight of 2.8 grams. The polymer was then dissolved into 140 ml of distilled water and stirred using a magnetic stirrer at 60 ± 1^{o}C with a stirring rate of 600 rpm. The solution was added Tween 20 as much as 0.8 g and garlic oil with a variety of variations as shown in Table 1. An alginate solution was obtained by dissolving 0.6 g na-alginate in 80 ml of water. The polymer solution was added dropwise into this solution. The mixture was stirred for 15 minutes while maintaining a constant temperature of 60°C. Then, the pH of the mixture was reduced to 3.75 by adding 2.5% (v/v) 98% acetic acid solution. After that, the solution was cooled until the temperature reached 5-10°C, and 0.183 grams of glutaraldehyde was added. Next, the temperature of the solution was raised again to 35°C and stirred for about 3 hours to complete the crosslinking reaction [6]. The solution with crosslinked particles was filtered using filter paper, then washed with distilled water. Then, the second stage of washing was carried out using an n-hexane to remove the oil attached to the surface of the microcapsule. After that, the microcapsule is dried to form a garlic oil microcapsule.

Table 1. Polymers and garlic oil composition

Sample	Gelatine (gr)	Maltodextrin (gr)	Garlic Oil (gr)
1	2,8	0	1
2	2,8	0	2
3	2,8	0	4
4	1,4	1,4	2
5	0	2,8	2

2.4 Result Analysis

The resulting microencapsulation was analyzed for microencapsulation yield, encapsulation efficiency, FTIR (Fourier Transform Infrared Spectrophotometer) and SEM (Scanning Electron Microscope) testing.

2.4.1 Yield of Microencapsulation (Yield)

Yield is used to determine the efficiency and effectiveness of a process. The higher the yield value, the more efficient the drying process will be. Several factors that affect the yield value are the microcapsule-forming material and the amount of essential oil added to the emulsion.

$$\text{Yield} = \frac{\text{Microcapsule Weight}}{\text{Microcapsule Forming Weight}} \times 100\% \quad (1)$$

2.4.2 Analysis with FTIR (Fourier Transform Infrared Spectrophotometer)

FTIR can detect functional groups as well as analyze mixtures of the samples being analyzed. In the microcapsule test, the results of the FTIR are in the form of a graph so that the composition contained in the sample can be analyzed through the peaks.

2.4.3 Analysis with SEM (Scanning Electron Microscope)

This tool can depict the surface of the material using an electron microscope with high magnification. The results of the SEM test are in the form of an enlarged image of the sample showing the microencapsulation texture in the sample.

2.4.3 Digital Microscope Analysis

Digital microcapsule analysis aims to take pictures of microcapsules and analyze the morphology of microcapsules. This digital microscope analysis was carried out using the CoolingTech Digital Microscope.

2.4.4 Encapsulation Efficiency

Encapsulation efficiency is determined by the principle of extraction using a volatile organic solvent such as petroleum ether. The extraction process is carried out by taking the required sample, then inserting the sample into the Soxhlet extractor. This test aims to calculate the volume of oil that was successfully encapsulated in the garlic oil coating process using gelatin polymer, maltodextrin, and sodium alginate.

$$\text{Encapsulation Efficiency} = \frac{\text{Extracted Oil}}{\text{Oil Added for Coating}} \text{ x } 100\% \quad (2)$$

3. RESULTS AND DISCUSSIONS

3.1 Microcapsule Yields

Yield is used to determine the efficiency and effectiveness of a process. The higher the yield value, the more efficient the encapsulation process is. The yield of microencapsulation is presented in Table 2. Table 2 shows that the lowest yield was found in sample 1 with 2.8 grams of gelatin polymer coating material and the amount of essential oil added was one gram (29.43%). A low yield value can indicate that the encapsulation process does not run efficiently. In the fifth sample using 2.8 maltodextrin coating, no microcapsules were produced. This happens because there are no polymer bonds that bind each other, so the oil cannot be encapsulated properly.

The addition of oil in microencapsulation has a significant effect on the resulting yield. The more the core material is added, the greater the resulting yield. This is estimated by the greater the core material covered, the greater the total solids produced. The high yield is supported by the greater amount of maltodextrin. The greater the amount of maltodextrin, the greater the total solids obtained so the yield value will be greater.

Table 2. The yield value of Garlic Oil microencapsulation.

Sample	Gelatine (gr)	Maltodextrin (gr)	*Garlic Oil* (gr)	Microcapsule Weight (gr)	Yield (%)
1	2,8	0	1	1,643	29,43
2	2,8	0	2	2,769	42,06
3	2,8	0	4	3,298	38,43
4	1,4	1,4	2	3,487	52,97
5	0	2,8	2	0	0

3.2 Analisis with FTIR

The FTIR spectra for the materials used can be seen in Figure 1. For garlic oil The 1634 cm^{-1} band was attributed to the C-C bonds in the allyl group of the garlic oil present in its vibrational stretching mode, while at a wavenumber of 915 cm^{-1} shows stretching vibrations in C-S-C bonds [11]. Maltodextrin has a peak at a wavelength of 1148 cm^{-1} and 1078 cm^{-1} which is C-O stretching and 992 cm^{-1} is a characterization of polysaccharides [12]. Gelatin has a peak at a wavelength of 1623 cm^{-1} and 1517 cm^{-1} which are amide I and amide II vibrations [13].

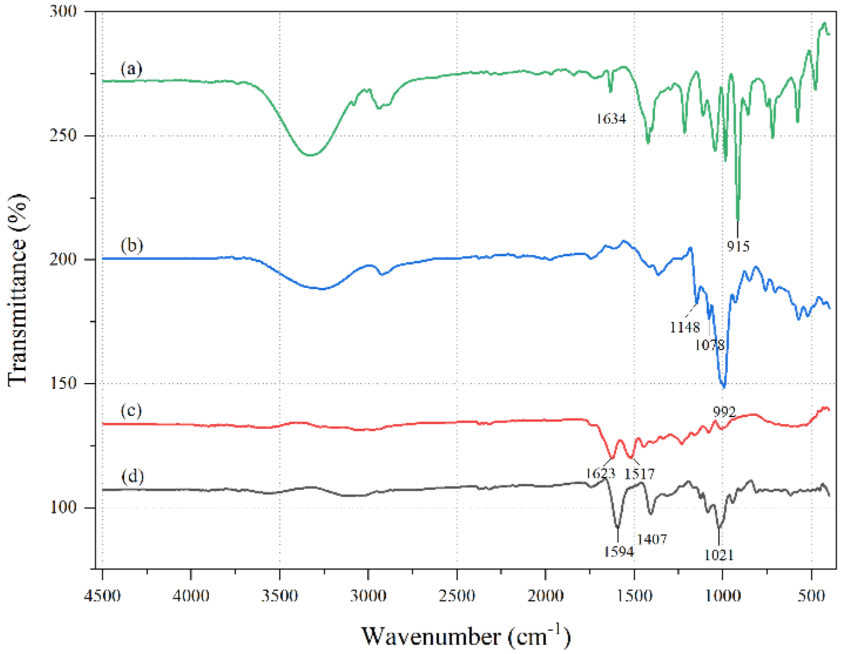


Figure 1. FTIR test results of the microencapsulation materials used; (a) Garlic oil, (b) maltodextrin, (c) gelatin, and (d) sodium alginate

The FTIR spectra for microencapsulated garlic oil are shown in Figure 2. The peaks at wavelengths of 1627 cm-1, 1633 cm-1, 1630 cm-1 and 1640 cm-1 indicating the presence of garlic oil with the allicin content in the resulting microcapsules.

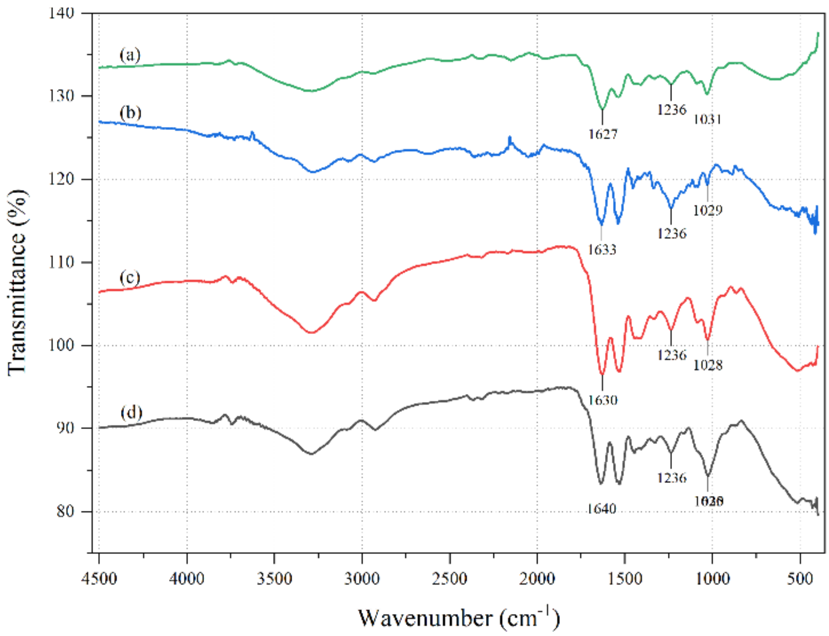
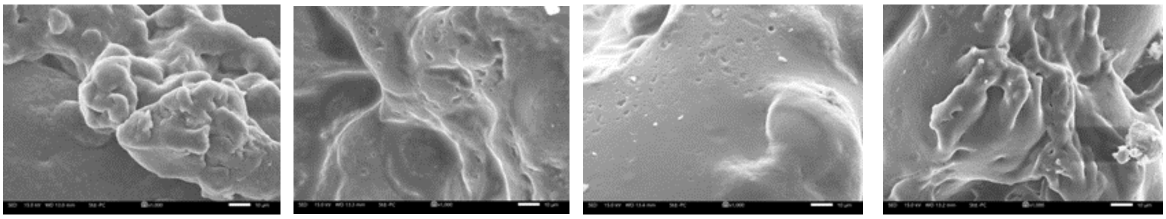


Figure 2. FTIR Test Results; (a) Sample 1, (b) Sample 2, (c) Sample 3, and (d) Sample 4

3.3 Analisis with SEM

The morphology of the microcapsules were analysis with its SEM images. In this SEM test, 1000x magnification was used for all samples. The SEM test was carried out on garlic essential oil microencapsulation samples with various variations as can be seen in figure (3). The results of the SEM test showed that the resulting microcapsules were asymmetrical and had an uneven surface. In figure (3) with a magnification of 1000x, it can be seen that sample 3 has the flattest surface compared to samples 1 and 2. This means that the sample with more oil composition will have a flatter surface. Meanwhile, sample 4, which was composed of 1.4-gram gelatin polymer, 1.4-gram maltodextrin, and 0.8-gram sodium alginate with 2-gram oil, had a very uneven surface morphology.

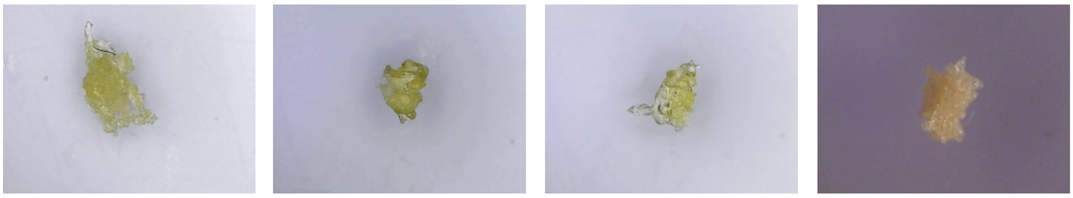


(a) (b) (c) (d)

Figure 3. SEM Test Results at 1000× magnification: (a) Sample 1, (b) Sample 2, (c) Sample 3, and (d) Sample 4

3.4 Digital Microscope Analysis

The microcapsule test with a digital microscope aims to analyze the morphological shape of the microcapsule. The results of the test with microcapsules can be seen in figure (4). The results of the digital microscope showed that the microcapsules were irregular and textured in shape.



(a) (b) (c) (d)

Figure 4. Digital Microscope Test Results (a) Sample 1, (b) Sample 2, (c) Sample 3, and (d) Sample 4

3.5 Encapsulation Efficiency

The amount of garlic oil that can be encapsulated during the microencapsulation process is analyzed by extracting it using n-hexane solvent, then drying it in an oven to remove the remaining solvent and water to obtain pure oil. As a result of the microcapsule extraction process, essential oils will be obtained. The essential oil is an oil that is encapsulated or coated in a matrix.

In Table 3, the data obtained from the encapsulation efficiency of Garlic Oil is presented, the highest efficiency value was obtained in sample 1 using 2.8 grams of gelatin polymer coating and the amount of essential oil added was 1 gram, which is 34.23%. The obtained encapsulation efficiency decreased with increasing the amount of essential oil added. The addition of maltodextrin as a solvent has the effect of increasing the yield value, but can also reduce the efficiency value. With the same amount of polymer and a different amount of oil added, it is possible that the polymer used is not sufficient so a lot of oil is not coated which resulting in a decrease in the efficiency of encapsulation.

Table 3. Garlic Oil Encapsulation Efficiency Value

Sample	Gelatine (gr)	Maltodextrin (gr)	*Garlic Oil* (gr)	Microcapsule Weight(gr)	Efficiency (%)
1	2,8	0	1	1,643	34,23
2	2,8	0	2	2,769	19,57
3	2,8	0	4	3,298	6,47
4	1,4	1,4	2	3,487	13,75
5	0	2,8	2	0	0

4. CONCLUSION

Garlic oil can be encapsulated using gelatin and maltodextrin with the coacervation method. The results obtained from SEM testing and digital microscopy showed that the resulting microcapsules were irregular in shape and had an uneven surface. The highest yield was in sample 4 with a composition of 1.4 grams of gelatin : 1.4 grams of maltodextrin, which was 52.967%. Increasing the amount of garlic oil can also cause a decrease in encapsulation efficiency. The highest encapsulation efficiency occurred in the addition of oil to sample 1 (1 gram) using 2.8 grams of gelatin coating, which was 34.23% with a yield of 29.43%.

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