The protection of fish gelatine and arabic gum as coating materials to the quality chlorophyll *Caulerpa racemosa* encapsulation

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Abstract. Chlorophyll from *Caulerpa racemosa* is a pigment that has the potential to be used as a natural coloring agent. However, chlorophyll is susceptible to high temperatures. Microencapsulation is a technology that can protect chlorophyll degradation. The purpose of this study was to determine the effect protection of fish gelatine and Arabic gum combination as a coating material to the quality of chlorophyll microencapsulation from *C. racemosa*. Five different combination formulations of coating materials composed by fish gelatine and Arabic gum at ratio (w/w) 0%:10% (A), 0.5%:9.5% (B), 1%:9% (C), 1.5%:8.5% (D), 2%:8% (E) were applied to chlorophyll microencapsulation process. The microcapsules quality were tested for yield, solubility, dissolved solids, bulk density, moisture content, chlorophyll levels, and a degree of lightness test. Other analyses such as FTIR, SEM and DSC confirmed availability of chlorophyll in microcapsules. The result showed that the formulation of C (combination of fish gelatine and Arabic gum at ratio (w/w) 1%:9%) has a high solubility of 97.75% and low water content of 3.57%, encapsulation efficiency of 68.95%, the density of 0.427 g/cm$^3$ and chlorophyll level of 18.46 mg/L. The presence of chlorophyll was indicated at wavelengths of 1583–1709, 2809–3012, and 3029–3639 nm$^{-1}$.

1. Introduction

*Caulerpa racemosa* (sea grape) is a green macroalga that grows in Indonesian waters and contains high chlorophyll levels (5.48 mg/g). Furthermore, the macroalgae contain chlorophyll a (5.48 mg/g), chlorophyll b (3.06 mg/g), and carotene (0.39 mg/g). High chlorophyll contents make *C. racemosa* a potential natural dye [1].

Synthetic dyes are becoming extensively used because they are inexpensive, and the color produced is attractive. Consumers avoid food products that used synthetic dyes because they are considered harmful to one’s health [2, 3]. Consumer preferences colour of food and drink now is moving to a natural product. One alternative choice for using natural coloring at tropical country is macroalgae *Caulerpa racemosa* since is widespread along the coastal region.

The photosynthetic pigments such as chlorophylls a and b are unstable to temperature and pH, so they are easily degraded into derivative molecules [4]. Chlorophyll quality is an ability of a product to maintain its properties and characteristics more or less about 50% when the chlorophyll was processed...
and storage. Chlorophyll quality is important to be maintained because chlorophyll is widely used in various industries such as food and pharmaceutical industries [5].

Microencapsulation is a process that protects and maintains active components that are easily degraded from environmental influences by using coating material as a protector. Microencapsulation includes coating particles with dimensions ranging from 1 µm to 5000 µm. The coating material used in the microencapsulation process must be non-toxic, unreactive with the core material, and adjustable to the desired end product. The coating materials commonly used for encapsulation are alginate, carrageenan, Arabic gum, gelatin, and maltodextrin. One reason for combining coating materials on encapsulation process are based on their successfully to protect the core material to release slowly.

Fish gelatin is a protein that can form gels, emulsifiers, and purifiers [6], whereas Arabic gum is a hydrocolloid produced by the tapping process of the Acacia tree, and there are texture formers binders, and emulsifiers [7]. The both two raw materials display good properties as a coating material, and both need to be combined to form a better microcapsule coating, hence the best concentration of both materials should be determined. The aim of the research was to determine the effect protection of combination formulation fish gelatine and Arabic gum as coating material to the quality of chlorophyll microencapsulation from C. racemosa as are followed yield, solubility, dissolved solids, bulk density, moisture content, chlorophyll levels, and a degree of lightness test. Analyses such as FTIR, SEM and DSC to be done in order to confirmed availability of chloropyll in microcapsules.

2. Methods

2.1. Material
Fresh C. racemosa seaweed samples were collected from Karimun Jawa Island, North Central Java, Province, Indonesia. The samples were dried in a shady place for 4–5 days to decrease the water content [8]. Fish gelatin are processed from tilapia bone that are supplied from tilapia fillet factory at Semarang, Arabic gum was prepared from Merck (Germany).

2.2. Chlorophyll extraction
Chlorophyll extraction was conducted following the procedure, and the ratio of solvent volume and extraction time was modified [9]. Fresh C. racemosa samples were cut to approximately 1 cm, weighed to 100 g, and crushed with a mortar. The samples were then placed in a glass bottle and extracted by immersion using 400 mL of ethanol solvent (Sigma Aldrich, USA) or a sample ratio with solvent, namely, 1:4 (v/v). The samples were macerated for approximately ± 48 h at room temperature, and then it was filtered paper to separate the filtrate with residue. The maceration filtrate were then evaporated with a rotary vacuum evaporator (Buchi Rotavapor R-200, Switzerland) at ± 20°C to form an extract that had no odor of solvent.

2.3. Chlorophyll microencapsulation
The microencapsulation process was carried out with the addition of 89% chlorophyll extract, 1% tween 80 (Sigma Aldrich, USA) and 10% coating material with the ratio (w/w) of fish gelatin : Arabic gum : 0%:10% (A); 0.5%:9.5% (B); 1%:9% (C); 1.5%:8.5% (D); and 2%:8% (E) into distilled water. The mixtures were homogenized using an homogenizer (WiseTis HG-15D, Germany) with a speed of 10,000 rpm for 3 min and then frozen at −35 °C for 24 h. The frozen samples were dried using a freeze dryer (Heto Powerdry LL 1500, Germany) at −100 °C vacuum pressure for ±48 h [10].

2.4. Yield
The yield was calculated by comparing the weight of the microcapsules obtained with the total active ingredients and coating components used [11]. The yield was calculated as follows:
Yield (%) = \frac{\text{weight of microcapsule biomass + coating component}}{\times 100\%}

2.5. Solubility
Approximately 0.5 g (a) of the samples was weighed, dissolved into 50 mL of distilled water, and filtered with a vacuum filter. Prior to use, filter paper was dried in a 105 °C oven (Binder, Germany) for about 30 min and then weighed (b). After the filtration process, the filter paper and residue of the material were dried in a 105 °C oven for 3 h, cooled in a desiccator for 15 min, and weighed (c). Solubility was calculated as follows [12]:

\[
\text{Solubility} (\%) = \left\{ 1 - \frac{c - b}{(100 - \%ka) \times a} \right\} \times 100\%
\]

2.6. Dissolved solid
The mixture of 0.5 g sample and 6 mL distilled water were homogenized and heated for 5 min, shaken, and heated in a boiling water bath (Memmert, Jerman) for 5 min. The mixture was cooled, added with distilled water to the level limit, and filtered. Subsequently, 5 mL of filtrate was obtained and placed in a porcelain dish (A). The filtrate was then evaporated to dryness using an oven at 105 °C to achieve a constant weight (B) [12].

\[
\text{DS} (\% \text{ db}) = \frac{(ab)}{b} \times (BA) / \text{dry weight sample x 10 } / 0.5 \times 100\%
\]

2.7. Bulk density
Bulk density was determined by dividing the weight of microcapsules with the volume of microcapsules [13]. The bulk density of samples of chlorophyll microcapsules was calculated by placing the microcapsules (5 g) in a measuring cylinder.

2.8. Moisture content
The sample was weighed (a), placed into a cup (b) to dry in an oven (Binder, Germany) at 105 °C–110 °C for 3–4 h cooled, and weighed (c) [12]. The moisture content was calculated as follows:

\[
\text{Moisture content} (\%) = \frac{(b - a) - (c - a)}{(b - a)} \times 100\%
\]

2.9. Chlorophyll content
Microcapsules were centrifuged at 1000 rpm for 10 min, and 1 mL of supernatant was diluted into a 10 mL measuring flask. The chlorophyll concentration was obtained from the absorbance measurements of extracts at 663 and 645 μm by a UV-Vis spectrophotometer (Shimadzu UV-1280, Japan). The chlorophyll concentration was calculated by using the following equation [9]:

\[
\text{Chlorophyll content} (\text{mg/L}) = (20,31. A_{645.0 \text{ nm}} + 8,05. A_{663.0 \text{ nm}})
\]

2.10. Encapsulation efficiency (EE)
Encapsulation efficiency was calculated by the ratio of the concentrate on of encapsulated chlorophyll with the chlorophyll concentration at the beginning process [14].

2.11. Color parameter (L*, a*, b*)
Twenty five grams chlorophyll microcapsules from *C. racemosa* transferred into a clear container and measured using a UV/VIS Color Measurement (Mettler Toledo Inc., Switzerland) in a temperature range of 25 °C–500 °C with a nitrogen speed of 100 mL/min. Measurements began when the indicator light was turned on. L, a*, and b* values were displayed on the screen [15].

2.12. Microcapsule function group

The chlorophyll functional groups in microcapsules were determined by Fourier transform infrared spectroscopy (Shimadzu, Japan) at 4000–400 nm [16].

2.13. Differential scanning calorimetry

Numerous microcapsules were analyzed by DSC (Mettler Toledo Inc., Schwerzenbach, Switzerland) in a temperature range of 25 °C–500 °C with a nitrogen speed of 100 mL/min [17].

2.14. Morphology

The morphology of microcapsules was observed by scanning electron microscopy (SEM; JEOL-JSM 6510LA, Japan) using a gold coating with a voltage of 10 kV [17].

3. Result and discussion

3.1. Yield

The yield obtained in chlorophyll microcapsules ranged from 47.21% to 51.41% (Table 1). The viscosity of the mixture material increases so that the mixture sticks to the drying container, which results in reduced microcapsule yields. Arabic gum can increase the viscosity and stability of a solution [18]. The yield of the product gradually increased when total solids were added to the mixture. This result was in accordance with the findings of Nogueira et al., [19], who reported that high amounts of coating material lead to high yields of microcapsules produced. Thus, the amount of coating material used is critical in rendering microcapsule products.

| Formula | Concentrate of Fish gelatin: Arabic gum | Yield (%) | Solubility (%) | Dissolved Solid (%) | Bulk Density (g/cm³) |
|---------|---------------------------------------|-----------|---------------|-------------------|---------------------|
| A       | 0%: 10%                               | 49.78 ± 0.99<sup>ab</sup> | 89.96 ± 1.54<sup>a</sup> | 95.67 ± 2.08<sup>b</sup> | 0.57 ± 0.01<sup>c</sup> |
| B       | 0.5%: 9.5%                            | 47.21 ± 2.22<sup>a</sup> | 91.79 ± 1.36<sup>a</sup> | 94.33 ± 2.08<sup>b</sup> | 0.53 ± 0.01<sup>d</sup> |
| C       | 1%: 9%                                | 50.38 ± 1.11<sup>ab</sup> | 97.75 ± 1.70<sup>b</sup> | 93.67 ± 1.52<sup>b</sup> | 0.43 ± 0.10<sup>c</sup> |
| D       | 1.5%: 8.5%                            | 51.41 ± 0.58<sup>b</sup> | 98.17 ± 1.05<sup>b</sup> | 92.33 ± 2.08<sup>b</sup> | 0.35 ± 0.01<sup>b</sup> |
| E       | 2%: 8%                                | 48.72 ± 1.18<sup>ab</sup> | 98.61 ± 0.89<sup>b</sup> | 85.67 ± 2.08<sup>a</sup> | 0.24 ± 0.01<sup>a</sup> |

Note:
- Data with different superscripts indicate significant differences (α = 0.05).
- Data are the mean of triplicate samples ± standard deviation.

3.2. Solubility

Table 1 showed the five microcapsule formulas produced relatively high solubility values ranging from 89.96% to 98.61%. Microcapsules should have high solubility, and it depended on the coating materials [2, 20]. Gelatin contains amino acids that are hydrophilic, so the gelatin has high water solubility and is suitable for use as a coating material because it can lead to high solubility. Arabic gum is an emulsifier, so any material added to it will easily dissolve in water. [21, 22]. High water content in the material caused the material to be difficult to spread in water because no pores were formed. As a result, the material was unable to absorb large amounts of water. Powders containing low water contents have a...
greater capacity to absorb water as a result of high hygroscopicity [23]. The increasing of the hygroscopic properties of microcapsules causes differences in water vapor pressure, so the higher the ability of particles to absorb water on its surface or the faster the microcapsules are moistened by water, the higher the solubility of microcapsules [24]. The encapsulation with freeze dryers produces products that are highly soluble in water because they have a high surface area (porous structure), hygroscopic type and easily rehydrated. The freeze-drying process occurs through a sublimation mechanism that occurs at cold temperatures because gelatinization, caramelization, and denaturation do not occur, and crust formation does not change in the dry part of food [25].

3.3. **Dissolved solid**
Dissolved solids measure the ability of chlorophyll microcapsules to dissolve in water. The test results on Table 1 demonstrated that the dissolved chlorophyll content from formula A microcapsules was not significantly different those from formulas B, C, and D but significantly different from that of formula E. The addition of 2% gelatin in formula D caused a decrease in dissolved solids.

3.4. **Bulk density**
An increase bulk density in microcapsule was associated with increased in Arabic gum concentration addition. The data from Table 1 was consistent with the findings of Mahdavi et al., [22], who found that bulk density of anthocyanin microcapsules increases by adding Arabic gum. The increase in bulk density is associated with moisture content. High moisture content increases bulk density [13]. Moreover, the amount of bulk density is related to the air inside the microcapsule.

3.5. **Moisture content**
Moisture content is a critical quality for microcapsules product since is associated with dissolved solid and storage life. During the encapsulation process, the hydroxyl group in fish gelatin will form hydrogen bonds with surrounding water molecules. If water is evaporated, crystallization will occur because the hydroxyl group will form linkage together to form monomers. Therefore, the higher the concentration of gelatin used, the faster the rate of crystallization and evaporation of water [26] so that the microcapsule dissolved solid (Table 1) and water content becomes lower. The addition of concentration fish gelatin more than 1% was not recommended since it caused sticky form of chlorophyll microcapsules. Arabic gum has a large molecular weight and a complex molecular structure so that the bond between molecules with water is strong [21]. When the freeze-drying process occurred, water molecules are difficult to evaporate from the microcapsules [27] since protein gelatin possed a stronger bond matrix.

| Formula | Concentrate of Fish gelatin: Arabic gum | Moisture Content (%) | Chlorophyll contents (mg/L) | Encapsulation Efficiency (%) |
|---------|----------------------------------------|----------------------|-----------------------------|-------------------------------|
| A       | 0%: 10%                                | 6.47 ± 0.17d         | 20.66 ± 0.31c               | 77.13±1.16b                  |
| B       | 0.5%: 9.5%                             | 5.19 ± 0.42c         | 19.27 ± 1.32c               | 71.97±4.93b                  |
| C       | 1%: 9%                                 | 3.57 ± 0.49b         | 18.46 ± 1.40bc              | 68.95±5.22b                  |
| D       | 1.5%: 8.5%                             | 1.90 ± 0.24a         | 15.13 ± 1.90b               | 56.51±7.09a                  |
| E       | 2%: 8%                                 | 1.06 ± 0.40a         | 13.64 ± 1.85a               | 50.95±6.91a                  |

Note:
- Data with different superscripts indicate significant differences (α = 0.05).
- Data are the mean of triplicate samples ± standard deviation
The result of moisture content shown in Table 2 was in line with Valentina et al., [28], who stated that the freeze-drying method could result in the moisture content of under 1%. In the freeze-drying process, crust not formed on the surface of the material so that water in the material can be evaporated properly [29]. Based on quality standard industry, microcapsules should contain moisture content below 5%. The regularly is regarding the shelf life of a product, which is influenced by humidity. Low moisture content will be likely to have a higher hygroscopic level. Microcapsule particles will absorb water quickly and caused the product to easily clump and affect the shelf-life [30, 31, 32]. According to data in Table 2, formula C has moisture content close to the industrial standard compared to other formulas.

3.6. Chlorophyll content

The chlorophyll content in C. racemosa prior to microencapsulation was 26.78 mg/L. After chlorophyll was encapsulated, there was a decreased in chlorophyll levels from formulas A to E. The chlorophyll contents were 22.87%, 28.03%, 31.05%, 43.68%, and 49.73%, respectively (Table 2). The addition of fish gelatin concentration was followed by a decreased in chlorophyll microcapsule levels. The acid properties of gelatin caused magnesium in chlorophyll to be displaced and replaced by two hydrogen atoms so that chlorophyll derivatives are formed, namely, pheophytin which has a pale green color [33, 34].

The decreased in chlorophyll levels in microcapsules formula A to formula E was still below 50%, which indicated that chlorophyll was not damaged during freeze drying process. Chlorophyll degradation in formulas A, B and C were significant different to that in formulas D and E, which were almost 50%. Therefore, the application of gelatin with a concentration of 1% to the formulation was a safe limit where chlorophyll was not degraded excessively. The chlorophyll a type was found 1.6776 ug/g dried basis, and there was not chlorophyll b was found, that mean chlorophyll was extracted with the proper solvent since the main type was expected was chlorophyll a.

3.7. Encapsulation efficiency (EE)

Chlorophyll microcapsules with fish gelatin and Arabic gum as coating materials was affect the encapsulation efficiency. The addition of 1% fish gelatin results in high encapsulation efficiency. This was indicated by the results of encapsulation efficiency of chlorophyll microcapsules in formula A not significantly different with formulas B and C but significantly different with formula D and E. The addition of concentration fish gelatin more than 1% decreased the encapsulation efficiency of microcapsules. This result is related to the chlorophyll content in microcapsules.

3.8. Color

In alkaline media (pH 7–9), chlorophyll is highly stable against seasoning but unstable in acidic media (pH 5) [35]. The redness value of the five formulas was negative, which indicated that the five formulas were green. Formulas A, B, and C produced greener microcapsules than formulas D and E (Table 3). Chlorophyll is included in the dark color so that the increase in chlorophyll levels will be inversely proportional to the value of L*. The results of measuring the brightness level are inversely proportional to the amount of chlorophyll, so an increase in the total amount of chlorophyll can reduce the brightness level of microcapsules.

| Formula | Concentrate of Fish gelatin: Arabic gum | L   | a       | b       |
|---------|----------------------------------------|-----|---------|---------|
| A       | 0%: 10%                                | 31.66 ± 1.53^a | -23.33 ± 0.58^a | 29.00 ± 1.00^a |
| B       | 0.5%: 9.5%                             | 33.33 ± 1.53^ab | -23.00 ± 1.00^a | 32.67 ± 0.58^b |
| C       | 1%: 9%                                 | 36.67 ± 2.08^b | -22.33 ± 0.58^ab | 35.00 ± 1.00^b |
### Table 3: Chlorophyll Microencapsulation by Freeze-Drying Method

| Formula | Fish Gelatin Concentration | Lightness (\(L^*\)) | Redness (\(a^*\)) | Greenness (\(b^*\)) |
|---------|-----------------------------|-----------------------|-------------------|---------------------|
| D       | 1.5%: 8.5%                  | 41.33 ± 0.58<sup>c</sup> | -20.67 ± 0.58<sup>bc</sup> | 39.33 ± 1.15<sup>c</sup> |
| E       | 2%: 8%                      | 42.67 ± 1.15<sup>c</sup> | -20.33 ± 0.58<sup>c</sup> | 41.67 ± 0.58<sup>c</sup> |

Note:
- Data with different superscripts indicate significant differences (\(\alpha = 0.05\)).
- Data are the mean of triplicate samples ± standard deviation.

There was a gradual increase of the lightness and red colour as increasing fish gelatin concentration addition. Chlorophyll microencapsulation of *C. racemosa* by freeze-drying method produced a green color. The data in Table 3 was supported by the study that the encapsulation process with the freeze-drying method will yield a homogeneous and good colour chlorophyll distribution throughout the encapsulation agent. The drying process with the freeze-drying method occurs at freezing temperatures of −100 °C so that Arabic gum does not show discoloration due to heating, which usually occurs during high temperature drying [36, 37]. Formula D and E have lighter yellow showed in higher L and b value than formulas A, B, and C. The microcapsules colour showed that the combination of coating materials in the freeze-drying process caused the green color of chlorophyll to leach off.

#### 3.9. Morphology microcapsules

The morphology of microcapsules by freeze drying were flake-shaped and it were related to the ability of the coating material to trap chlorophyll (Figure 1). Although in the form of amorphous surface flakes, the structure of microcapsules could encapsulate chlorophyll efficiently. The same research on oleoresin has a similar amorphous flake-shaped structure [38]. Research on polyphenol microcapsules showed the same pattern there was a shaped structure like broken glass through freeze drying method [39].

#### 3.10. Differential scanning calorimetry

The DSC thermograms of microcapsules in Figure 2 showed an interaction between chlorophyll and coating material, namely, fish gelatin and Arabic gum. Figure 2 A, B, C and E showed more than one temperature peak appeared for chlorophyll microcapsules of formulas A, B, D, and E. Formula C (Figure 2C) had only one peak temperature, which indicated an success interaction between the encapsulated coating and chlorophyll material as a core. Furthermore fish gelatin, Arabic gum and chlorophyll were able to interact well, and chlorophyll was perfectly entrapped in the coating material. The same results were obtained in curcumin microcapsules, where one peak temperature in curcumin microcapsules, which indicated an interaction between curcumin and coating material [40]. The other study on ascorbic acid encapsulation showed a similar pattern with one peak temperature [41].
**Figure 1.** Microstructure of chlorophyll microcapsules (the ratio of fish gelatin:arabic gum: 0%:10% (A); 0.5%:9.5% (B); 1%:9% (C); 1.5%:8.5% (D); and 2%:8% (E), fish gelatin (GEL); Arabic gum (GA).
Figure 2. DSC Thermogram of chlorophyll microcapsules (the ratio of fish gelatin:Arabic gum:0%:10%(A); 0.5%:9.5%(B); 1%:9%(C); 1.5%:8.5% (D); and 2%:8%(E), fish gelatin (GEL), arabic gum (GA)).

Figure 3. FTIR Spectra of Chlorophyll Microcapsule (nm\(^{-1}\)) (The ratio of fish gelatin:Arabic gum:0%:10%(A); 0.5%:9.5%(B); 1%:9%(C); 1.5%:8.5% (D); and 2%:8%(E), fish gelatin (GEL), arabic gum (GA), chlorophyll (CHLO)).

3.11. Microcapsule functional groups
The results of FTIR analysis demonstrated that all chlorophyll functional groups were detected after finishing the microencapsulation process (Figure 3). Hence, chlorophyll was able to be encapsulated with fish gelatin and Arabic gum coating. The presence of chlorophyll was indicated by functional groups C=O aldehydes, CH groups, and OH groups at wavelengths of 1583–1709, 2809–3012, and 3029–3639 nm\(^{-1}\), respectively [42]. In this study, each functional group was detected at wavelengths of 1626 nm\(^{-1}\), 2927 nm\(^{-1}\), and 3386 nm\(^{-1}\). Similar results were obtained by Kumar et al., [43], who reported wavelengths for C = O groups, CH groups, and OH groups at 1625.8, 2852.0, and 3381.3 nm\(^{-1}\), respectively.

4. Conclusions
The differences in the combination of coating material affect the yield, solubility, water content, bulk density, chlorophyll level, and color intensity of chlorophyll C. racemosa microcapsules. Formulas A
and B have more than 5% of moisture content, which would affect the shelf-life. Formula C with a combination of fish gelatin: Arabic gum (1:9% w/w) produces the best quality with a yield of 50.38%, solubility of 97.75%, the water content of 3.57%, bulk density of 0.427 g/cm³, chlorophyll level of 18.46 mg/L, and values of L* of 36.67, a* of 22.33, and b* of 35.

Conflict of interest
All authors declare that they have no conflict of interest.

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