HASIL CEK_Capsule shells from Eucheuma Cottonii Seaweed With Plasticizer Sorbitol and Filler TiO2

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Capsule shells From Eucheuma Cottonii Seaweed With Plasticizer Sorbitol And Filler TiO2

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***Abstract*—Most of capsule shells commercial production recently from gelatin as raw material, but the application of gelatin in hard capsule shells production can causes controversy. That is the emergence of consumers concern about health problem and religion issues.Total production of seaweed in the world reached 30.1 million ton per year that makes Indonesia became second position as the biggest producer of seaweed in the world that produce 11,631 tons per year. The species of seaweed that used in this research is Eucheuma cottonii as raw material to make capsule shells because it contains carrageenan, that act as product stability regulator, thickening, gelling agent, emulsifier and as bioplastic fibers in capsule shells which contains cellulose 20.62%. The elastic properties can be obtained from the addition of sorbitol as plasticizer and TiO2 as filler. The variation with making capsule shells and ends with sample testing. Then the sample will be analyzed for water content, ash content, solubility of capsule shells in water, uniformity of weight and acidity (pH).**

Keywords—capsule shells, seaweed, sorbitol, TiO2

I. Introduction

The various kind of medicine is consumed by human to keep their health. But there are some kind of medicine when consumed have a bitter taste or aroma that cause human doesn't want to consume it. So to solve this problem a solid preparation is made, which is called a capsule shells. Capsules shell can devided into hard capsules shell and soft capsules shell. Commonly, hard capsules shell is produced from gelatin, known as Hard Gelatine Capsules (HGC). It generally consists of 80 to 85 percent gelatin and about 10 to 15 percent water. The largest source of gelatin right now is from pig and cow. Reference [1] present that the total production of gelatin in the world reach 326,000 tons per year. Based on data, the percentage of gelatin from pigskin about 46%, cowhide about 29,4%, cow bone about 23,1% and other source about 1,5%.

Furthermore, gelatin from pig have cheaper price than cow gelatin, so that many capsule shells producers are more inclined to use the gelatin from pig [2] The price of pig gelatin is about US$ 5,67 while cow gelatin is about US$ 14,17 [3].

Use of gelatin for pharmaceutical needs estimated around 17% from the consumtion of gelatin in the world [4]. In Indonesia until now gelatin is still an imported product, based on the Ministry of Industry on 2017 noted that gelatin that used in pharmaceutical industry increased from 2015-2016 with precentage 7.51%. With import value on the 2015 about US$ 1.861.900 and on the 2016 about US$ 2.001.800.

Use of gelatin in capsule shells production causes controversy, such as issues with BSE, religious dietary restrictions with pork-based products, consumer preferences for healthy veggie products, and animal rights concerns have led to the demand for nonanimal-based capsule shellss [5]. Bovine Spongiform Encephalopathy (BSE) or mad cow disease scare in 1990s. This issue triggered a FDA program to scrutinize the use of animal-derived materials in manufacture of pharmaceutical products. Every FDA filing requires certification, which in turn requires suppliers properly certify that their animal-derived raw materials have minimum risk BSE and Transmissible Spongiform Encephalopathy (TSE) [6]. And from pig is feared because it might contains swine flu disease. Religion issues that appear such as hinduist forbidden to consume cow, moslem and jewish forbidden to consume all products that contain pig.

This condition encourage people to find the alternative which is to replace the raw material capsule shells production that is gelatin with a material which can be applied in process to make capsule shells. One of the efforts to solve the problem is use base material that is not from the animal but with the plant-based material that still has the good compatibility properties like gelatin [2]. One of the alternative is using nongelatinous polymers, such as hypromellose (hydroxypropyl methylcellulose [HPMC]) and strach. The originally invented HPMC capsule shellss contain a secondary gelling agent such as kappa-carrageenan or gellan. Carrageenan are algal (seaweed) polysaccharides consisting of high-molecular weight linear sulfated galactan chains. [6]. Seaweed are one of the abundant material in Indonesia, and this potency can be used to make capsule shells. This paper presents our research in making capsule shells from seaweed to replace capsule shells from gelatin. Seaweed also contains high fiber, and it is celar that the fiber content of algae (seaweed) is a high as in terrestrial plants or even higer [6].

The overall objectives of this research are :

- To make halal capsule shells from seaweed as raw material, so it safe for consumption.
- To explore potential of seaweed as alternative to replace gelatin in process making capsule shells.
- To eliminate contoversy about capsule shellss from gelatin.
- This research was conducted to compare with capsules made from gelatin.

II. Literature Review

Indonesia has a sea area of 70%, wherein there are various types of marine biota in it. One of the biota that the cultivation must be developed these days is seaweed. The high potential of Indonesian seaweed to be developed is not only because economically seaweed has a high economic value, but more importantly Indonesia is an archipelago with an untapped potential planting area of 50%. The total potential of seaweed planting area that still available is 769.5 thousand hectares [8]. According to data from reference [9], since 2010-2016 seaweed production in Indonesia has increased.

Thus placing Indonesia at the second position at the largest seaweed producer in the world with a total production of 38.7%, and in the first position is China with a total production of 47.9%.

Eucheuma cottonii is a type of seaweed that used as an alternative basic material of capsule shells. This seaweed is chosen because it has the highest carrageenan content among the other types. Carrageenan is a polysaccharide extracted from the red seaweed from Rhodophyceae class which can be used as raw material for polysaccharide-based products, one of which is a hard capsule shells. Based on [10], the species Eucheuma cottoni produced the highest percentage of carrageenan content compared to the species Eucheuma sp. such as Eucheuma spinosum, and Eucheuma edule. The percentage of carrageenan content for Eucheuma cottonii was 61.54%, Eucheuma spinosum was 46.64%, and Eucheuma edule was 54.56%. The very significant difference shown in the analysis of the variety of carrageenan content is caused by different gel strength in each species, where Eucheuma cottonii with stereotyped content structures in the form of carrageenan kappa, Eucheuma spinosum with structural stereotypes of carrageenan iota, and Eucheuma edule with uterine stereotypes in the form of carrageenan lamda. Indonesia is the largest producer of carrageenan-producing seaweed in the world, with a quantity of 3399 thousand tons with a percentage of 60.5% from the total production in the world [11].

In the making process of capsule shellss, plasticizers must be added so that the resulting capsule shells is more elastic, flexible, and resistant to water. In this research, the plasticizer we use was sorbitol. This addition aims to improve the physical properties, mechanical properties, and protect plastic from microorganisms that can damage it. Other than that, TiO2 is added as a filler or bleaching agent in capsules. Titanium dioxide is very stable at high temperatures, white in colour, amorphous, tasteless, and not hygroscopic [8] [12]. Capsule colouring material is using food gade colouring such as Tartrazine Cl 19140 and Ponceau 4R Cl16255. Characterization of commercial carrageenan and FAO standards.

TABLE I. TABLE OF CHARACTERIZATION OF COMMERCIAL CARRAGEENAN

Carrageenan	Specification			
	Water (%)	*Ash (%)*	*Sulphate (%)*	*Viscosity (cP)*
Standard	<12	15-40	15-40	>5

a. Source : FAO, 2007

A. MATERIALS AND METHODS

1. Equipment

Laboratory tools obtained from the Laboratory of unit process Chemical Engineering Departement, Faculty of Industrial Technology, Ahmad Dahlan University. Such as Oven, beaker glasses, capsule mold, glass stirrers, dropper pipes, hot plates, thermometers, digital scales, watch glasses, stopwatch, water bath, motor stirer.

2. Ingredients

Carrageenan from Eucheuma cottoni seaweed, TiO2, distilled water, colouring (tartazin and ponceau 4R), sorbitol.

TABLE II. TABLE OF VARIABLE USED IN THE RESEARCH UNITS IN WEIGHT RATIO IN GRAM

Variable	Variable (label)			
	A	*B*	*C*	*D*
Carrageenan	3	4	5	6

Aquadest which had been heated until 60℃ with stirring during 3 minute. During stirring added sorbitol to the solution. Put the solution in water bath which has been regulated temperature is 800C. The solution will be placed in a water bath for 45 minutes. Next, the capsule mold is dipped into the mixture before the mixture begins to harden. The result of the mold dried in an oven, to reduce the water content, at 40℃ for 5 hours.

3. Procedure

1 gr of TiO2 and weight variation of Eucheuma cottonii carrageenan (3g, 4g, 5g, and 6g) dillute with 60 ml.

Where :

A = Weight of evaporating dish (g)
B = Weight of evaporating dish with sample (g)
C = Weight of evaporating dish with ash (g).

- Uniformity of Weigth
  Weight analysis is done by weighing the capsule shells weight and then analyzing the suitability of commercial capsule shells weight set at 69-83 mg/100 capsule shellss [13].
- Analysis of Solubility in Water
  This analysis is quite easy, by inserting the sample into each graduated cylinder that contains distilled water. The temperature of distilled water used is 37 °C. After that, observe the time needed for the sample to dissolve in the distilled water. Water resistance for commercial capsule shellss set by reference [13] is minimum of more than 15 minutes.
- Analysis of Acidity
  The analysis of acidity (pH) is done by mashing the sample, then the mashed sample is weighed as much as 2 g. After weighing, dissolve the sample with 20 ml of distilled water. Stir until everything dissolves then analyze the solution using pH meter.

4. Analysis of Capsule shells

In this research samples that have been made will be tested. There are 5 types of the analysis which will be done to find out the best sample from the four samples that have been made. That is analysis of water content, ash content, solubility in water, uniformity of weight and pH. For uniformity of weight analysis will be tested at Chemical Engineering Laboratory Ahmad Dahlan University. And for the others samples were tested in Pharmaceutical Laboratory of Ahmad Dahlan University.

- Analysis of Water Content (AOAC 2005)

  Analysis of water content is done by evaporation using an oven. According to AOAC (Association of Official Analytical Chemist) in 2005 stated analysis of water content stage. The first step is dried evaporating dish in the oven at 105 °C for 1 hour. Next, the evaporating dish placed into a desiccator (approximately 15 minutes) and left until it is cold then weigh it. After that, put the sample into evaporating dish and oven with temperature of 102-105 °C for 5-6 hours. Then put evaporating dish into the desiccator and let it cool (30 minutes), weigh it. Calculation analysis of water content in capsule shellss is :

$$\% \text{ Water content} = \frac{B-C}{B-A} \times 100\% \qquad (1)$$

Where :
A = Weight of evaporating dish (g)
B = Weight of evaporating dish with sample (g)
C = Weight of evaporating dish with dried sample (g)

- Analysis Of Ash Content (AOAC 2005)

The principle of the analysis of ash content is to determine the amount of ash contained in a material related to minerals from the analyzed material. The evaporating dish is cleaned and dried in an oven at around 105 °C for 30 minutes. Then put evaporating dish into a desiccator (30 minutes) and weigh it. Next, put the sample into evaporating dish, then weigh it. After that, burn evaporating dish and the sample in a furnace with temperature of 600 °C for 7 hours. Put the evaporating dish in the desiccator, let it cool and weigh it. Calculation analysis of ash content in capsule shellss is :

$$\% \text{ Ash content} = \frac{C-A}{B-A} \times 100\% \qquad (2)$$

B. RESULT AND DISCUSSION

1. *Analysis of Water Content (AOWC 2005)*

The results of the analysis of water content capsule shells from carrageenan are presented in Image 1. The results of the analysis shows that the water content of the capsule has a value 25.53 - 31.82 %. The water content of a product usually determined by the conditions of drying, packaging and storage methods [14]. In figure 1, the relation between carrageenan weight and the percentage of water content is as follow.

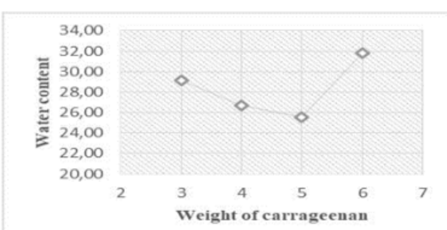

Fig. 1. Correlation between weight carragenan and percentage of water content

Based on figure 1 we can conclude that the best water content percentage is in the sample of C treatment using 5 g carrageenan with the result 25,53%. Then followed by sample of B treatment with value equal to 26,67%. The ratio of carrageenan weight between B and C treatment is 4:5. This provide that the more carrageenan used will decrease the water content of capsule shells. But actually at the time of making capsule shellss, there are some dependent variables which very influential, that is temperature and the use of sorbitol. In D treatment although the weight of carrageenan used was higher than C treatment, the water content of D treatment was higher and even higher than the other three samples. It is causes by the temperature and drying time, because in the D treatment the solution is viscous so that the resulting capsule shells is thicker.

The capsule shells's water content in this research is bigger than the comercial capsule shells. Capsule shells is a product made from organic material and usually will be overgown with fungis and molds if the water content is more than 20%. That's why according to the data from reference [13], the specification of capsule shells's water content is ranging between 12,5% - 15 %. So that water content will affect storage time from the capsule shells.

2. *Analysis of Ash Content (AOAC 2005)*

Ash in the capsule shells is an anorganic material, like minerals in the comercial capsule shells, must have a small ash content. Based on Indonesian Ministry of Health, 1995, ash content in a capsule shells no more than 5% . The results of ash content from carrageenan are presented in figure 2:

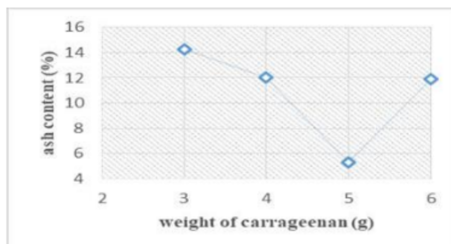

Fig. 2. Correlation between weight of carrageenan and percentage of ash content

Based on figure 2. we can conclude that the best ash content percentage is on capsule shells labeled with C (5,26%) then followed with B (12%). The capsule shells that labeled B that with the treatment 60 ml of distillated water, 5 ml sorbitol as plasticizer, 5 g of carrageenan, and 1 g of TiO2.

The variabels that affect the value of ash content are almost the same as the variables that influence the analysis of the water content.

3. *Uniformity of Weight*

Weight analysis of the capsule shells aims to find out the thickness of the capsule shells. The thicker of capsule shells, then the weight of capsule shells is increases. The capsule shells weight is one of the standard, which must be fulfilled for commercial capsule shellss. The weight of capsule shells commercial is ranging between 69-83 mg/100 capsule shellss [13]. On figure 3, have been presented the results of uniformity of weight analysis.

Fig. 3. Correlation between weight of carrageenan and weight of 100 capsule shells

Based on figure 3, we can conclude that carrageenan capsule shells with C treatment has the result value according with capsule shells commercial. It shows that the bigger carrageenan used so the result value of weight of 100 capsule shells is getting bigger too.

The factors that influence result of capsule shells analysis are weight of carrageenan, sorbitol, drying temperature, and drying time. The use of a little carrageenan can makes the product has a thinner thickness. Addition of sorbitol as a plasticizer can increase flexibility and permeability of water vapor, gas and increase resistance especially if stored at low temperatures. The drying temperature and drying time very influential on the final product because if it is too dry, the product will be difficult to remove from the mold. And for a thick capsule shells solution, the product that will be produced is dryer.

4. Analysis of Solubility In Water

The capsule shells serves as a packaging for drugs in the form of powder. The capsule shells which is easily damaged or easily penetrated by water can cause drug that inside of capsule shells dissolved, so that the bitter taste will be tastes. The water resistance for commercial capsule shellss as determined by reference [13] is more than 15 minutes. From the analysis results it has been found that there is a correlation between the weight of carrageenan and the disintegation time of capsule shellss in water on figure 4

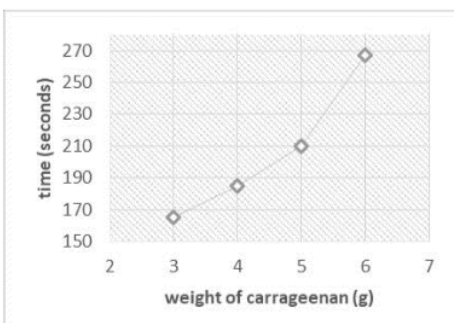

Fig. 4. Correlation between weight of carrageenan and time of solubility in water.

Based on the data above, we can conclude that the solubility of capsule shellss in all samples accord the specifications, that is more than 15 minutes. However, the more weight the carrageenan that used the more disintegration time needed. That's because of the thickness of the capsule shells. And also the effect of sorbitol used. Like the function of sorbitol, which is as a plasticizer so with adding sorbitol in the manufacture of capsule shellss will affect the disintegration time of solubility in the water.

5. Analysis of Acidity

One of the commercial capsule shells parameters is acidity (pH). According to the Indonesian Ministry of Health in 1995, the acidity (pH) of capsule shellss that must be fulfilled is in the range of 5 to 7. On figure 5, have been presented the results of acidity (pH) from sample that have been produced:

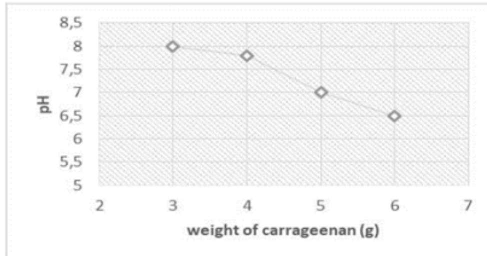

Fig. 5. Correlation between weight of carrageenan and acidity (pH)

Based on figure 5. The results of C and D treatment accordance with the acidity of capsule shells commercial. It shows that the smaller carrageenan used so the result value of acidity is getting bigger.

From the research that has been done, the product in the form of capsule shellss with the results presented in figure 6.a; 6.b; 6.c; and 6.d.

Fig. 6.a 3g carrageenan

Fig. 6.b 4g carrageenan

Fig. 6.c 5g carrageenan

Fig. 6.d 6g carrageenan

III. CONCLUSION

In this study can be conclude that capsule shells can be made from carragenan that contained in Eucheuma Cottoni seaweed. In making capsules shell sorbitol is used as plasticizer and TiO2 is used as filler. Sample C has the best value of water content, ash content, and acidity (pH). With a value of 25, 53% for water content, 5.26% for ash content and 7 for the acidity (pH). For ash content analysis B trearment fulfills the parameters of the commercial capsule shells standard. Determination of C treatment as the best mixture of ingedients. For the solubility test in water A treatment is the best results because it dissolves faster than the other samples. Where the time needed is 165 minutes. Fixed variables also very influential in testing, such as the addition of sorbitol, drying time, and drying temperature.

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