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Encapsulation of Lemongrass Extract (*Cymbopogon citratus*) Coated Alginate/Chitosan Using Gelation Method

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Abstract

Lemongrass (Cymbopogon citratus) is a medicinal plant with various biological activities such as antibacterial, antifungal, antiprotozoal, anti-inflammatory, and antioxidant. This study aimed to encapsulate lemongrass bioactive in alginate/chitosan complex by enhancing the properties of $CaCl_2$ crosslinked incorporated with tween 80 by ionic gelation method. The hydrogel was prepared by mixing alginate solution (2% w/v) and chitosan solution (1% w/v) with a ratio (1:1 v/v). Tween 80 (2% v/v) was added as a dissolution enhancer and $CaCl_2$ as a crosslinker agent. The formulation varying by concentration of $CaCl_2$ (0.1M to 0.3M) and lemongrass extract (2% to 12%). Encapsulation lemongrass with alginate-chitosan beads is characterized to determine encapsulation efficiency, swelling study, morphology, functional groups, and release study. The results showed that encapsulation efficiency ranged from 74.81% to 83.07%. Encapsulation efficiency increased with the addition of $CaCl_2$ and lemongrass extract concentration. The swelling ratio ranged from 27.29 to 37.81, it will decrease with the addition of $CaCl_2$ and lemongrass extract concentration. The Scanning Electron Microscopy (SEM) analysis of hydrogel beads shows a polyhedral shape, porous, and rough surface which indicates bioactive of lemongrass trapped on the beads. The Fourier Transform Infrared Spectroscopy (FTIR) results show new peaks at 1734 cm^{-1} as carbonyl stretch vibrations in ketones, aldehydes, and carboxylic acids, indicating the addition of lemongrass extract. Bioactive of lemongrass extract loaded alginate-chitosan beads was successfully released as much as 87.12% at pH 6.8. This study suggested the strong potential alginate-chitosan beads as an encapsulating agent for lemongrass extract using the ionic gelation method, and it has potential as a drug delivery system.

Keywords: *encapsulation; lemongrass; alginate; chitosan; $CaCl_2$*

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INTRODUCTION

The medicinal plant is defined as a type of plant that is partially or whole parts of the plant are used as medicine, cosmetics, and health. It is claimed to improve body immunity because it contains secondary metabolites (Olorunnisola *et al.*, 2014). Based on the Ministry of Agriculture Republic of Indonesia on the Central Bureau of Statistics (2016), 15 types of medicinal plants are cultivated in Indonesia, whereas lemongrass is not included. However, lemongrass has much biological activity because it's essential oils and phenolic compounds (Haque *et al.*, 2018). Lemongrass *(Cymbopogon citratus)* is an aromatic plant that is widely cultivated in Central and South America, most of Africa, Southeast Asia, and the Indian Ocean Islands (Majewska *et al.*, 2019). Lemongrass has also been known to control platelet composition, cure diabetes, gastrointestinal infections, tract digestibility, anxiety or depression, malaria, and pneumonia. Whereas in the industrial, they use as additives, flavors, and preservatives in beverages and food (Oladeji *et al.*, 2019). It mentioned because of their biological activity in the lemongrass as an antibacterial, antifungal, antiprotozoal, anti-inflammatory, antioxidant, antitussive, antiseptic, anticarcinogenic, and antirheumatic (Ekpenyong *et al.*, 2015), antidiabetic (Bharti *et al.*, 2013), insecticide (Oladeji *et al.*, 2019), and anti-dermatotoxicity (Carmo *et al.*, 2013).

Lemongrass is usually consumed for herbal treatment with conventional brewing as liquid form, such as lemongrass tea. However, liquid forms has a short shelf life, impracticable, and lacks economic value than powder form. Therefore, technology to manipulate lemongrass is needed. It has to be practical but does not reduce the quality of lemongrass, encapsulation is the breakthrough solution (Akbari-Alavijeh *et al*, 2020). Some studies had previously been done, such as encapsulation of leaf tea green extract (Zokti *et al.*, 2016), guava extract leaves (Tuan *et al.*, 2016), and stevia extract leaves (Arriola *et al.*, 2019). However, encapsulation of lemongrass extract is still rarely performed, so it becomes a breakthrough. Lemongrass powder is already processed through crystallization (Estiasih *et al*, 2017). Whereas lemongrass contains anti-diabetic substances which diabetics cannot consume if it processing by crystallization (Bharti *et al.*, 2013). Accordingly, encapsulation is a suitable method for drug delivery systems. Encapsulation is a common technique for creating an external layer or coating of one material over another material, which is applied for protection and preservation of bioactive that volatile also easily biochemical and thermal degradation. It is also used to mask the taste and aroma which is undesirable, and coated are used in lemongrass encapsulation is alginate/chitosan (Saifullah *et al.*, 2019).

Alginate is a hydrocolloid anionic polymer extracted from brown algae that have become the most common material for encapsulation (Atencio *et al.*, 2020). Alginate is used as a material of encapsulation due to its biocompatibility and nontoxic material in food (Sreekumar and Bindhu, 2019). The addition of chitosan to the encapsulation provides a double advantage, improving the nutritional and functional properties of the material. Chitosan is a linear polysaccharide that is slightly hydrophilic (Raza *et al.*, 2020). Furthermore, chitosan has amino and hydroxyl groups that charged positively and created cationic biopolymers soluble in water (Hamed *et al.*, 2016). Previously research has shown that direct mixing encapsulation with alginate/chitosan has some weaknesses. It damages the morphological structure of the material considering the difficult homogeneity between alginate and chitosan itself (Nalini *et al.*, 2019; Figueiredo *et al.*, 2020). In this research, the ionic gelation method is used with Tween 80 as an emulsifier to mix alginate and chitosan. $CaCl_2$ is used as an alginate crosslinker agent, forming beads and improving the material's physical and mechanical properties. This method has advantages, the beads effectively protect the bioactive, increase stability, increase encapsulation efficiency, and improve the properties of particles are generated (Figueiredo *et al.*, 2020). Several researchers were studied of lemongrass encapsulation. Melo *et al.*,(2020), have developed a lemongrass encapsulation using maltodextrin by spray drying. Erminawati *et al.*, (2019), have studied lemongrass encapsulation using maltodextrin and β-cyclodextrin by spray drying. Encapsulation of lemongrass extract in alginate-chitosan is still rarely performed. Therefore, this research was to investigate the effect of $CaCl_2$ and lemongrass extract concentration on lemongrass encapsulation with encapsulation efficiency, swelling study, SEM, FTIR, and release study.

MATERIALS AND METHOD

Materials

Lemongrass extract was obtained from PT. Intan Chemical, Surabaya, Indonesia, with specification relative density at 20/20 °C: 0,87-0,89 and carbonyl compounds content (minimum 75%). The chemical materials such as sodium alginate (molar mass 216.12 g/mol with CAS Number 9005-38-3 SIGMA-Aldrich, USA), chitosan (75-85% degree of deacetylation with CAS Number 9012-76-4 SIGMA-Aldrich, USA), calcium chloride ($CaCl_2$), tween 80, acetic acid (CH_3COOH), hydrochloric acid (HCl), sodium hydroxide (NaOH), and buffer solution (Merck Chemical Co., Darmstadt, Hesse, Germany). All chemicals used analytical grade.

Preparation of Blank Alginate-Chitosan Bead

Blank alginate-chitosan beads were prepared by dissolving 2% (w/v) sodium alginate using distilled water and homogenized with a magnetic stirrer for 30 minutes at 28ºC. Chitosan solution prepared by dissolving 1% (w/v) chitosan in dilute acetic acid solution and homogenized using a magnetic stirrer for 30 minutes at 40ºC. Dilute acetic acid is prepared by dissolving 96% acetic acid as much as 0.96 grams in 1000 mL of distilled water. After that, a solution of alginate and chitosan with a 1:1 ratio is mixed for 15 minutes until homogeneous. After stirring, the alginate-chitosan solution was dripped with a syringe into the $CaCl_2$ solution with concentrations of 0.2 M. The mixture will form a hydrogel in the $CaCl_2$ solution, wait for 30 minutes. Filter the hydrogel and then dry at 30ºC for 48 hours.

Lemongrass Encapsulation

Encapsulation method adapted from Camacho *et al.*, (2019) with some modifications. Alginate-chitosan solution was prepared as in the previous point. After that, alginate-chitosan solution was added with tween 80 2% (v/v) and lemongrass extract (2%, 4%, 6%, 8%, 10%, and 12% v/v) to the mixture. Lemongrass extract was obtained from taking lemongrass juice by refining and filtering using Whatman number 42 filter paper. Then the mixed solution was homogenized using a magnetic stirrer for approximately 10 minutes. After homogeneous, the mixture was dripped with a syringe into $CaCl_2$ solution with various concentrations (0.1 M; 0.15 M; 0.2 M; 0.25 M; and 0.3 M). The mixture will form a hydrogel in the $CaCl_2$ solution for 30 minutes. Filter the hydrogel and then dry at 30ºC for 48 hours.

Swelling Study Measurements

The swelling percentage was calculated by finding the average swelling percentage of hydrogels. The swelling test was determined using buffer solution at pH 7. Dried beads (0.2 gram) were diluted on a 20 ml buffer solution for 30 minutes. The swelling percentage calculated using the equation:

$$Swelling\ Study = \frac{W_s - W_d}{W_d} \quad (1)$$

Ws is the weight of the swollen beads and *Wd* is the weight of the dried beads (Ozbilenler *et al*., 2020).

Encapsulation Efficiency

This test was carried out to determine the amount of bioactive that was trapped in the alginate-chitosan matrix. This measurement was evaluated by indirect method. 2 ml of $CaCl_2$ solution was taken and then 2 mL of $AlCl_3$ and 2 mL of KCH_3COO were added. After that, the bioactive content was determined using the SP-300 Optima Spectrophotometer at a wavelength of 365 nm. Encapsulation efficiency is calculated using the following equation (Cirri *et al.*, 2021):

$$\%EE = \frac{Qt - Qr}{Qt} \times 100\% \quad (2)$$

Qt is the amount of bioactive present in the lemongrass extract and Qr is bioactive of the lemongrass extract present in $CaCl_2$ solution after encapsulation.

SEM Analysis

The morphological results of encapsulation of lemongrass with alginate-chitosan characterized using a Scanning Electron Microscope (SEM) JEOL JSM-6510LA

FTIR analysis

Fourier Transform Infrared Spectroscopy (FTIR) spectrum analysis was carried out for unloaded and lemongrass extract loaded alginate-hitosan beads using the KBr pellet method on the Perkin Elmer Spectrum IR 10.6.1 spectrophotometer (Perkin Elmer Inc., US) to confirm chemical bond information or functional group. The spectrum was recorded in the range of 4000-450 cm^{-1}.

Encapsulation Release Study

The release study was conducted in pH 2 and 6.8 solutions. Encapsulated lemongrass extract (0.5 gram) was dissolved in 30 ml of each solution and soaked for 24 hours. Samples were prepared in separate beakers per increment of time. For each time interval, 5 mL of sample was taken to be analyzed with SP-300 Optima spectrophotometer at a wavelength of 365 nm to determine the release of bioactive from lemongrass extract at any time.

RESULTS AND DISCUSSION

Swelling Study

Hydrogel is a material formed due to crosslinking between polymer chains and crosslinking agents. In this research, the alginate-chitosan hydrogel is formed by a physical crosslinking agent, $CaCl_2$. As a matrix constituent, alginate-chitosan has low mechanical and high degradation which $CaCl_2$ as a crosslinking agent can improve the properties of the material to form the hydrogel. Alginate-chitosan beads are formed due to a reaction between anion of carboxyl groups on alginate and cation of amino groups on chitosan. Figure 1 shows the interaction between alginate and chitosan, which can form a good complex due to opposite charges and interact with each other to form a 3D porous structure scaffold (Kumbhar and Pawar, 2017). The addition of ions Ca^{2+} of $CaCl_2$ to alginate-chitosan polymer causes the binding of two G chains in the alginate on the side of the opposite. Thus, the bond is formed a hole-shaped diamond consisting of a cavity hydrophilic that binds to ions of Ca^{2+} with coordination using atomic oxygen from the group carboxyl form the "egg-box" that shown in Figure 1. Furthermore, the crosslinking gives the structure of a 3D network, so the beads do not dissolve and allows the effective release of bioactive (Maitra and Shukla, 2014).

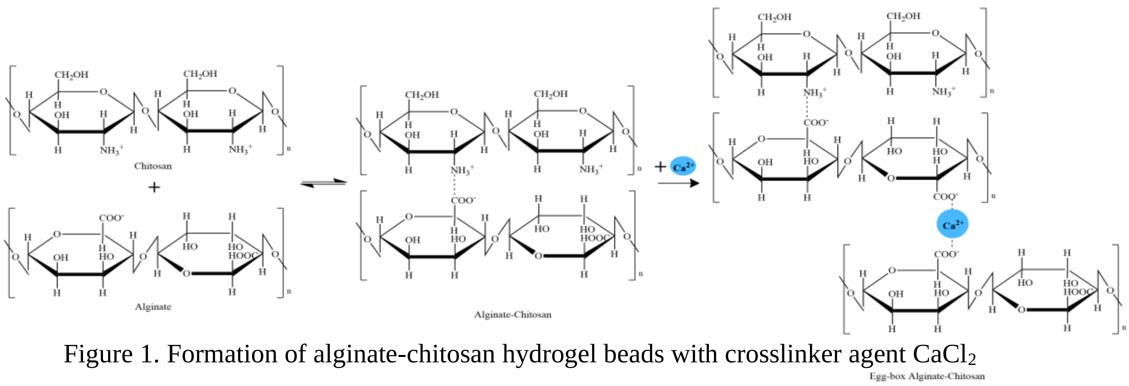

Figure 1. Formation of alginate-chitosan hydrogel beads with crosslinker agent $CaCl_2$

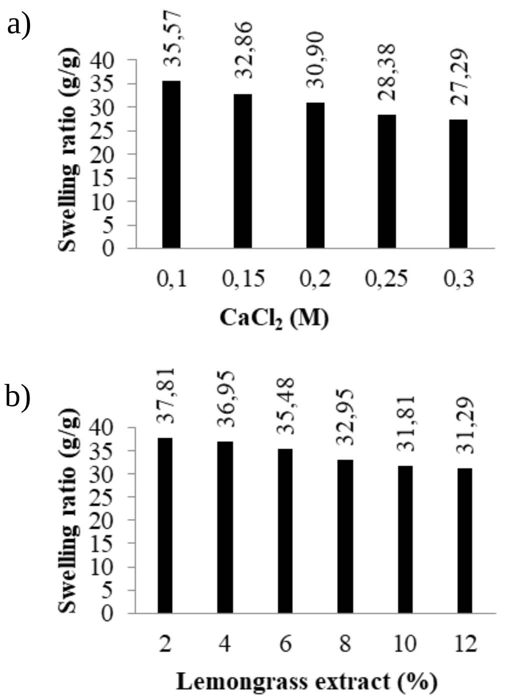

Figure 2**.** Effect of (a) $CaCl_2$ concentration (b) Lemongrass extract on swelling ratio

The hydrogel swelling will decrease with the addition of $CaCl_2$. As shown in figure 2 (a), $CaCl_2$ concentration from 0.1 to 0.3 M resulting in a swelling ratio of 35.57 to 27.29. The swelling concentration of 0.1 M is more substantial than 0.3 M. Increasing $CaCl_2$ concentration will produce stronger Ca^{2+} bonds, making the hydrogel more difficult to swell (Hariyadi *et al.*, 2014). In contrast, the effect of lemongrass extracts concentrations against the swelling ratio, shown in figure 2(b). At lemongrass extract concentrations 2-12% resulted in a 37.81-31.29 swelling ratio. The swelling ratio decreased along with the amount of lemongrass loaded. Hydrogel with low loading lemongrass has some groups hydrophilic are abundant as group hydroxyl and amino groups, which contribute to water absorption. However, increasing loading of lemongrass in the hydrogel may occupy most of the structure of space hydrogel, which will restrict the capacity of water absorption so the swelling and degradation is low (Chen *et al.*, 2019). Qi *et al.*, (2020) and Wang *et al.*, (2019) obtained similar results, increasing bioactive loading will reduce the value of the swelling ratio. Another research was conducted by Wu et al. (2018), which generates alginate-chitosan matrix with a swelling ratio of 30-42 containing lysozyme 0-2 mg / mL.

Encapsulation Efficiency

Encapsulation efficiency determines the levels of bioactive compounds in lemongrass extract entrapped in alginate-chitosan beads. Effects of $CaCl_2$ and lemongrass extract at various concentrations are shown in Table 1. As a result, adding $CaCl_2$ concentration can raise the encapsulation efficiency.

Table 1. Encapsulation Efficiency at various variables

Variable	$CaCl_2$ (M)	Lemongrass extract (% v/v)	EE (%)
1	0,1	5	74,81
2	0,15	5	76,24
3	0,2	5	77,49
4	0,25	5	78,20
5	0,3	5	78,90
6	0,2	2	80,21
7	0,2	4	81,70
8	0,2	6	81,82
9	0,2	8	82,10
10	0,2	10	82,67
11	0,2	12	83,07

The lowest encapsulation efficiency occurs at $CaCl_2$ concentration 0.1 M is 74.81%, and the highest at 0.3 M amounted to 78.9%. When $CaCl_2$ concentration increase, the availability of ions Ca^{2+} also increase, which cause a stronger cross link of the alginate-chitosan matrix, thus providing more bioactive compounds in lemongrass extract trapped (Gulati *et al.*, 2011). The study has been carried out by Hariyadi et al. (2014), $CaCl_2$ concentration, 0.1 to 1.5 M, has obtained an encapsulation efficiency of 49-88%. (Yousefi *et al.*, 2020) also carried out $CaCl_2$ 0.6-1.5 M concentration resulted in encapsulation efficiency of 67-77%. It is similar to this study, increased $CaCl_2$ concentration improves encapsulation efficiency.

Encapsulation efficiency of hydrogel also increases with the addition of lemongrass extract. According to Guo *et al.*, (2018), increasing bioactive concentrations would improve encapsulation efficiency, although not too significant. In the study, encapsulation efficiency was obtained in the range of 74 -83 % and it can be known that the addition of lemongrass extract 12% has highest encapsulation efficiency is 83.07 %. Due to the lemongrass extract concentration followed by exact matrix that potentially increased encapsulation efficiency. Previous research carried out by Fathordoobady *et al.*, (2021) showed that encapsulation betacyanin of dragon fruit peel extracts using alginate beads resulted in of 78.62%. Another study shown encapsulation of vitamin C using chitosan beads produce an encapsulation efficiency of 70% (Alishahi *et al.*, 2011). Increased encapsulation efficiency also indicates that delivery throughout the human body can be improved by reducing the losses before forming the gel. However, the concentration of 12% still can be absorbed in the human body (Guo *et al.*, 2018).

Scanning Electron Microscopy Analysis

The surface morphology of alginate chitosan beads by SEM shown in figure 3.

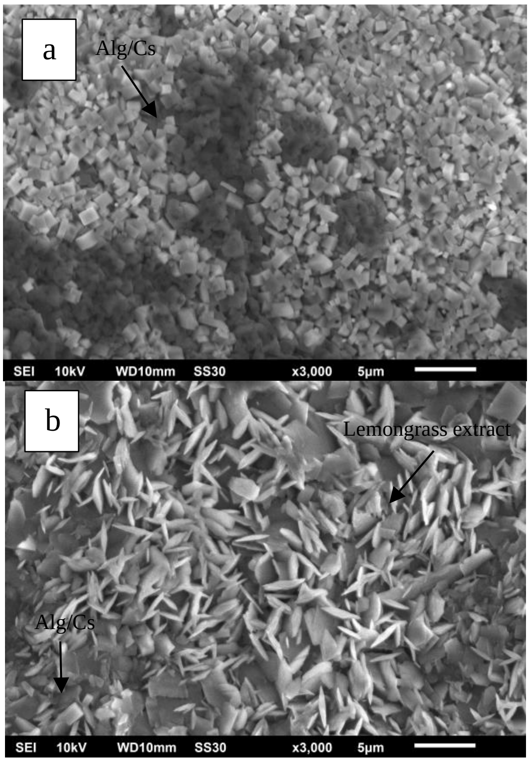

Figure 3. Surface morphology (a) blank alginate-chitosan matrix (b) alginate-chitosan matrix with 12% lemongrass extract

Figure 3(a) shows the surface morphology of alginate chitosan beads with crosslinking agent $CaCl_2$ 0.2 M, it looks microstructure and non-homogeneous with polyhedrical particles. $CaCl_2$ bond with alginates makes the cubic's morphological matrix but chitosan makes the morphology changes into polyhedrical (Daemi and Barikani, 2012).

Figure 3 (b) is a lemongrass-loaded alginate chitosan matrix with crosslinker agent $CaCl_2$ 0.2 M and Tween 80 2%. It is shown that the matrix surface is rough. These alginate-chitosan rough surfaces indicate that lemongrass bioactive is successfully trapped into the matrix (Vidart *et al.*, 2018). This result is similar to Lacerda et al., (2014) that the addition of rifampin causes the alginate-chitosan surface to become rougher. The study of Maniecka *et al.* (2016) also showed that the encapsulation of esculin with alginate-chitosan microparticles resulted in a rough surface. Furthermore, the surface morphology of the alginate-chitosan matrix containing lemongrass extract is porous. Therefore, it can support the release of lemongrass bioactive through the pores.

Fourier Transform Infrared Spectroscopy Analysis

FTIR analysis is shown in figure 4. Figure 4 (a) showed a spectrum of alginate-chitosan matrix with crosslinker agent CaCl2 0.2 M. At the peak absorption of 3331.94 cm-1, N-H stretching vibration is visible and overlaps with OH stretching vibration.

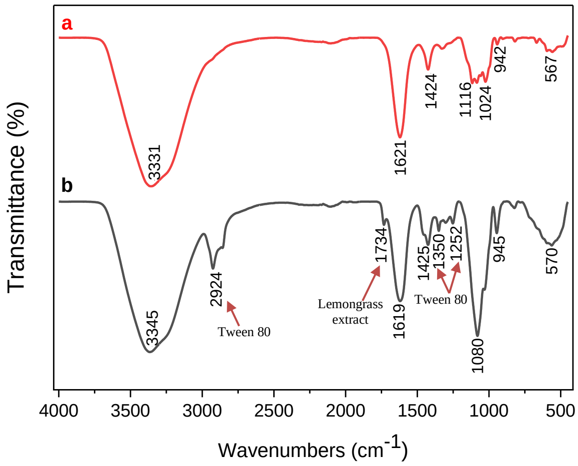

Figure 4. FTIR spectrum on (a) blank alginate-chitosan matrix (b) alginate-chitosan matrix with 12% lemongrass extract obtained from infrared absorption in the range of 4000–450 cm^{-1}

Furthermore, at 1621.24 cm^{-1} and 1116.43 cm^{-1}, in each peak seen that there is C = O vibration strain and O-H torsion vibration acid carboxylic and C-O stretching vibration. It is appropriate by the previous study (Lim and Ahmad, 2017; Velandia *et al.*, 2020). Peak at 1424.79 cm^{-1}, overlapping of symmetric COO- stretching and OH bending that indicate Ca^{2+} as crosslinker agent during alginate-chitosan crosslinking (Pilipenko *et al.*, 2019). The absorption peaks of alginate-chitosan beads were also shown at 942.59 cm^{-1} and 567.36 $cm^{-1,}$ indicating HC=CH and NH bending vibrations (Khan *et al.*, 2019).

Furthermore, FTIR spectrum of lemongrass loaded alginate chitosan matrix with crosslinker agent $CaCl_2$ 0.2 M and Tween 80 2% is shown in figure 4(b). The results are shown in several peaks which almost has an equal value. Several new peaks were found due to the loading of lemongrass and tween 80 addition. The lemongrass extract show the absorption bands at 1734 cm^{-1}. It is characteristic of carbonyl stretch vibrations in ketones, aldehydes, and carboxylic acids (Agarwal *et al.*, 2018). Tween 80 shows some absorption at 1252.42 cm^{-1}, 1350.19 cm^{-1}, and 2924.61 cm^{-1}. At 1350.19 cm^{-1} representing of C–H groups, especially –CH at the hydrocarbon chain (Javani *et al.*, 2021). The peak at 1252.42 cm^{-1} is associated with functional group of ester and ether (C-O) on aromatic ring and hydrocarbon chain, also the absorption peak at 2924.61 cm^{-1} represents a group of alkane C-H in long chain hydrocarbon (Molina *et al.*, 2014; Javani *et al.*, 2021). Besides that, the peak 1024.33 to 1080.47 cm^{-1} indicates the complex of alginate-chitosan with lemongrass extract was good inter-material constituent encapsulation and lemongrass bioactive successfully encapsulated in the matrix of alginate-chitosan (Antoniraj *et al.*, 2020).

Release Study of Encapsulation

The release profile of the bioactive of lemongrass extract from alginate-chitosan polymer at

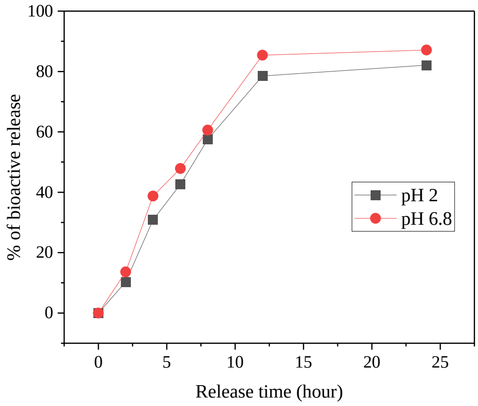

Figure 5. Release profiles bioactive extracts of lemongrass in pH 2 and 6.8 from encapsulated 12% lemongrass extract with $CaCl_2$ 0.2 M at alginate chitosan beads

various times is shown in Figure 5. Release studies were determined at pH of 2 and 6.8, as the representation simulated gastric fluid and simulated intestinal fluid. Only 4% of bioactive lemongrass extracts successfully release in the first hour, followed by rapid release up to 87% in 24 hours. This increase in the drug release may be attributed to the fact that the drug that is bioactive of lemongrass extract adsorbed may be concentrated on the core encapsulation (Nalini *et al.*, 2019).

The release of the bioactive lemongrass extract more rapidly at pH 6.8 than pH 2. Effect of pH associated with the state of ionization group and amino group carboxylate in the whole structure of chitosan and alginate. In an acidic solution (pH<6.5), the concentration of H^+ in the solution is high. Ion H^+ enters the network microcapsules and neutralizes the force of repulsion electrostatic. It causes a low relaxation rate so that the rate of release of the bioactive lemongrass extract becomes slow (Lim and Ahmad, 2017).

At pH> 6.5, amine groups are deprotonated and starting to reject the group carboxylic negatively charged so that the bond between amine and carboxylate group becomes weak. As a result, the release of bioactive from alginate chitosan beads becomes more quickly (Lim and Ahmad, 2017). The study is similar to Nalini *et al.* (2019) that the release of quercetin from nanoparticles of alginate chitosan more quickly occurs at pH 7.4 than pH 5.5 and 6.5. Another study also showed that encapsulation viola-odorata in alginate chitosan beads indicated low release at pH of 1.5 and higher release at 7 (Yousefi *et al.*, 2020). This study showed that the bioactives of lemongrass extract were protected in acidic media and released at pH 6.8.

CONCLUSION

Lemongrass bioactive extract can be encapsulated with an alginate-chitosan matrix. The results showed that encapsulation efficiency ranged from 74.81% to 83.07%, and the swelling ratio ranged from 27.29 to 37.81. SEM analysis showed that the lemongrass-loaded alginate chitosan beads have rough and porous surfaces. The FTIR result indicates successful loading lemongrass extract into alginate chitosan beads. Release study showed controlled release in acidic solution and higher release rate at pH 6.8. This study suggested the strong potential alginate-chitosan beads as an encapsulating agent for lemongrass extract using ionic gelation method and it has potential as drug delivery system.

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