Microencapsulation of red ginger oleoresin in maltodextrin and carrageenan using spray drying

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Abstract

The purpose of this study was to determine the encapsulation efficiency and cumulative release of red ginger oleoresin from microcapsules with different wall materials. Red ginger oleoresin was added to the maltodextrin solution, followed by a tween 80. The mixture formed was transferred to a spray dryer for the drying process. Other materials used are carrageenan and a combination of maltodextrin and carrageenan in a ratio of 1:1, 2:1, and 1:2. Red ginger oleoresin microcapsules were analyzed for encapsulation efficiency and release test using phosphate buffer medium pH 7.4, then determine release kinetics using zero-order, first-order, Higuchi model, Korsmeyer-Peppas model, and Peppas-Shalin model. The highest encapsulation efficiency was 78.6%, and the lowest cumulative was 58.46% from m.3, 6% dan kumulatif nakan medium buffer f. The release kinetics best fit the Korsmeyer-Peppas and Peppas-Shalin models with anomalous transport (non-Fickian) and Fickian diffusion release mechanisms.

Keywords:
Maltodextrin, carrageenan, red ginger oleoresin, spray drying.

1. Introduction

Ginger (Zingiber officinale Rosc.) is mostly used as food preservatives and medicine. It is widely used in China, Indian, and Arab to treat several diseases such as inflammation, rheumatics, stroke and diabetes [1-2]. Red ginger (Zingiber officinale var. rubrum) is part of ginger varieties with small rhizomes with larger phenol and flavonoid compound than white ginger [3]. Red ginger contains active components such as gingerol, shogaol, and zingeribene, often used as antioxidant [4], antibacterial [5], anti-inflammatory [6], and anti-cancer [7].

Ginger contains essential oil and oleoresins. The bioactive compound in ginger oleoresin is more than in the essential oil because the oleoresins have volatile and non-volatile compounds [8]. The main compounds from ginger oleoresins are gingerol, shogaol, zingerone, and paradol [9]. The weaknesses of red ginger oleoresins are sensitive to light, heat, and oxygen. Thus, the bioactive component can be degraded, resulting in an unpleasant taste and odor.
that reduce the possibility to direct consumption [10-11]. Microencapsulation is a useful method to protect the active ingredient from degradation. This process is used in the food and fragrance industry to protect essential oils, oleoresins, flavour and aroma [12].

Microencapsulation is a technique of coating active ingredients using a thin layer as a wall material. This method could be applied to protect the core material from the external environment (UV radiation, heat, and other factors), remove unpleasant odour and taste, reduce the speed of losses and evaporation of the core material, produce a solid form that is easier to handle, and provide controlled release of the active product [13]. The selection of the microencapsulation process depends on the core and wall material's chemical and physical properties [14]. Spray drying, freeze-drying, extrusion, fluidized bed, and emulsion crosslinking have been developed for the microencapsulation process [15]. Spray drying is one of the physical methods most widely used in the food industry [12-13].

The function of wall materials is to protect the core material from external that cause degradation, to reduce the reactivity of the core material, reduce evaporation, and control the release of the core materials. The wall materials can be derived from carbohydrates, proteins, and gums [16]. In this context, it is very important to compare the effect of different materials on the properties of microcapsules. Maltodextrin is commonly used as the wall material in the microencapsulation of food ingredients. It offers advantages such as low cost, high solubility, neutral aroma and taste, and good protection of flavours against oxidation. Carrageenan is a natural polysaccharide extracted from red seaweed. The nature of kappa-carrageenan can form a firmer gel than cornstarch and gelatin. Carrageenan also increases the retention time of the aroma and slow its release [17]. This study used maltodextrin and carrageenan with various combinations as a coating material for producing microencapsulated red ginger oleoresin by the spray-drying method. The encapsulation efficiency and the amount of oleoresin released were characterized.

2. Research Methodology

2.1. Microencapsulation of Red Ginger Oleoresin

The materials used in this study were red ginger oleoresin (obtained from Lansida group), maltodextrin and carrageenan as coating material, ethyl alcohol and n-hexane as a solvent to analyze surface oil and total oil, and Tween 80 as surfactant. The encapsulation process was prepared using spray drying by dissolving 20g of maltodextrin with 1000 mL aqua dest. About 20 g of red ginger oleoresin was added to the maltodextrin solution, followed by 2% Tween 80 from the total volume. The mixture was stirred using an overhead stirrer for 15 minutes to form the emulsion. The emulsion of red ginger oleoresin was then placed in the feed tank of a spray dryer at 120°C and 2 bars. The spray drying process is carried out by heating the material with maltodextrin walls.

2.2. Encapsulation Efficiency (% EE)

2.2.1. Surface Oil

The suspension was then filtered and dried using an oven. Dried microcapsules (1g) were added to 15 ml n-hexane and stirred for 1 min to dissolve surface oil. The amount of surface oil was calculated by the difference between initial and final weight after washing.

2.2.2. Total Oil

The total oil was determined using a soxhlet extraction unit. About 2g of oleoresin microcapsules were extracted using ethanol as a solvent for 6 hours to ensure complete oil extraction. The powder residue was dried and weighed. The encapsulation efficiency was calculated based on surface oil and total oil. The method was a modification from [18-22]. The equation used to determine encapsulation efficiency is as follow.

\[
\text{Surface oil} = \text{initial weight} - \text{final weight of microcapsules}
\] (1)

\[
\text{Total oil} = \text{initial weight} - \text{weight after extraction using soxhlet}
\] (2)

\[
\% \text{Encapsulation efficiency} = \frac{\text{Total oil} - \text{Surface oil}}{\text{Total oil}} \times 100
\] (3)

2.3. Cumulative Release Analysis

The release method of red ginger oleoresin from microcapsules with coating material maltodextrin, carrageenan and their mixture were prepared by [23] with modification. One gram dried red ginger oleoresin microcapsules soaked in buffer phosphate and stirred with speed 150rpm at 37±0.5°C. The sample was taken every 15 minutes interval until 180 minutes and then analyzed using spectrophotometer UV-Vis (Thermo scientific Genesys 10 UV) with the wavelength of 283 nm. The cumulative release values were used to calculate the release kinetics using five mathematical methods [24-25] as follow.

1. Zero order

\[
\frac{M_t}{M_{\infty}} = k_0 t
\] (4)

2. First order

\[
\frac{M_t}{M_{\infty}} = a[1 - \exp(-bt)]
\] (5)
3. Higuchi model

\[
\frac{M_t}{M_\infty} = k_H t^{1/2}
\]  

(6)

4. Korsmeyer-Peppas model

\[
\frac{M_t}{M_\infty} = k_{K-P} t^n
\]  

(7)

5. Peppas-Sahlin model

\[
\frac{M_t}{M_\infty} = k_1 t^m + k_2 t^{2m},
\]  

(8)

where \( \frac{M_t}{M_\infty} \) is red ginger oleoresin cumulative release, \( k_a, (a \text{ and } b) \), \( k_{H}, k_{K-P}, (k_1 \text{ and } k_2) \) are constants from zero order, first order, Higuchi model, Korsmeyer-Peppas model, Peppas-Sahlin model, \( n \) indicated release mechanism, and \( m \) is the diffusional exponent.

3. Result and Discussion

3.1. The Encapsulation Efficiency

The encapsulation efficiency is used to determine the ability of wall material for coating the bioactive compound properly. The values of encapsulation efficiency were calculated based on surface oil and total oil in equation 3. Less amount of oleoresin on the surface of microcapsules shows more efficient encapsulation process.

In Figure 1, high encapsulation efficiency was 60% at carrageenan wall material, and low encapsulation efficiency was 50% at maltodextrin material. Carrageenan can increase emulsion stability. [26] also stated that the addition of carrageenan could form a thick and stable emulsion. The high viscosity of carrageenan caused the red ginger oleoresin to be coated and increased the drying process, reducing the red ginger oleoresin diffusion to the surface. Encapsulation efficiency is affected by the amount of surface oil and total oil. Surface oil is the amount of oil on the surface of the microcapsules that are not coated. The low surface oil indicates that the encapsulation process is successfully done. Total oil affects encapsulation efficiency because increasing coated oleoresin will increase encapsulation efficiency.

The encapsulation efficiency of red ginger microcapsules using maltodextrin as wall material is smaller than the previous research by [12], which is 64.7% - 82.1%. In this study, the encapsulation efficiency was low because the maltodextrin concentration used at 2% (w/v) was also lower than the previous research. The concentration of wall material affects the retention of the bioactive compound in the feed solution. A high concentration of wall material can increase the retention of bioactive compounds due to the decrease in the drying time in the spray dryer. A low concentration of maltodextrin may reduce the ability for coating the oleoresin and makes the oleoresin diffuse and evaporate to the droplet surface during the drying process.

![Figure 1. The influence of difference wall material toward encapsulation efficiency of red ginger oleoresin](image1)

![Figure 2. The influence of maltodextrine and carrageenan ratio as wall material to the encapsulation efficiency of red ginger oleoresin.](image2)

The modified mixture of maltodextrin and carrageenan with different weight ratios gives different encapsulation efficiency of red ginger oleoresin microcapsule. Combining these materials increase the encapsulation efficiency, although it is not significant. The encapsulation efficiency of microcapsule...
using maltodextrin and carrageenan at weight ratio 1:1 is lower than at weight ratios 1:2 and 2:1. In Figure 2, the high encapsulation efficiency is 78.6% at a weight ratio of 2:1. The value of encapsulation efficiency from red ginger oleoresin microcapsules with maltodextrin or carrageenan as wall material were lower than the red ginger oleoresin microcapsules with various combination of maltodextrin and carrageenan as wall material.

Maltodextrin as wall material has low emulsion stability that results from a weak skin layer. Thus, another biopolymer will be essential to improve the strength of the wall material, such as carrageenan [27]. Figure 2 shows that the higher carrageenan concentration at a weight ratio of 1:2 could decrease the encapsulation efficiency. Meanwhile, the encapsulation efficiency increased when the amount of maltodextrin was added. The addition of carrageenan increased the moisture content of microcapsules [27]. Increasing moisture content resulted in a longer drying time. Hence the amount of coated oleoresin was reduced, and oleoresin, which was not coated, would evaporate during the spray drying process.

3.2. Cumulative Release

Wall material type and ratio significantly affect the cumulative release. Figure 3 shows the cumulative release of red ginger oleoresin from microcapsules. In Figure 3, the highest cumulative release, 94.49%, resulted from the mixture of maltodextrin-carrageenan at ratio 1:1, the lowest one, 58.46%, obtained from the mixture of maltodextrin-carrageenan at ratio 1:2. The time-release in this study was 180 minutes. The highest cumulative release can be achieved by using plain maltodextrin as wall material and maltodextrin–carrageenan ratio 1:1. The high release of red ginger oleoresin was caused by its high solubility [28].

The cumulative release of red ginger oleoresin from microcapsule with maltodextrin wall material was higher than microcapsule using combined wall material due to the high concentration of red ginger oleoresin at the microcapsule surface. Increased surface oil would decrease the encapsulation efficiency. Cumulative releases of red ginger oleoresin from microcapsule using carrageenan as wall material and combined material with a high concentration of carrageenan were low due to a thick and stable emulsion when carrageenan was added [26].

![Figure 3. Cumulative release profile of red ginger oleoresin from microcapsules using different types of wall materials](image)

3.3. Release Kinetics

The controlled release profiles are used to (1) prolong the drug release from dosage forms and reduce the adverse effect, (2) enhance bioavailability and reduce dose frequency for improved patient compliance. Release kinetics describes the overall drug release because quantitative and qualitative changes can alter drug release and performance in vivo. Release kinetics model such as zero-order describes the dissolution of the drug from the dosage form that does not separate and release the drug slowly. First-order models are used to describe drug absorption or elimination. The Higuchi model should not analyze release with the swelling system. However, this model is widely used for polymer systems that can expand because the model is simple. The release mechanism can be described with Korsmeyer-Peppas or Peppas-Shalin models.

| Wall material                  | Release kinetics model | Zero order | First order | Higuchi | Korsmeyer-Peppas | Peppas-Shalin |
|-------------------------------|------------------------|------------|-------------|---------|------------------|---------------|
|                               |                        | $k_0$      | $R^2$       | $a$     | $b$              | $R^2$         | $k_1$        | $k_2$        | $m$          | $R^2$       |
| Maltodextrine 2% (w/v)        |                        | 0.561      | 0.95        | 91.315  | 0.013            | 0.99          | 6.379        | 0.99         | 6.429        | 0.491       | 0.99         | 5.110        | 0.273       | 0.99         |
| Carrageenan 2% (w/v)          |                        | 0.370      | 0.96        | 64.450  | 0.011            | 0.97          | 4.190        | 0.98         | 6.254        | 0.451       | 0.98         | 2.504        | 0.295       | 0.98         |
| Maltodextrine: Carrageenan (1:1) |                      | 0.686      | 0.89        | 102.369 | 0.017            | 0.99          | 7.871        | 0.97         | 6.622        | 0.512       | 0.97         | 8.482        | 0.242       | 0.97         |
| Maltodextrine: Carrageenan (2:1) |                      | 0.317      | 0.97        | 110.285 | 0.004            | 0.97          | 3.544        | 0.95         | 1.134        | 0.739       | 0.97         | 1.138        | 0.369       | 0.97         |
| Maltodextrine: Carrageenan (1:2) |                      | 0.587      | 0.77        | 73.238  | 0.056            | 0.97          | 6.922        | 0.92         | 29.507       | 0.191       | 0.99         | 29.501       | 0.096       | 0.99         |
The value of n can determine the release mechanism of the drug. Ritger and Peppas determined the value of n and the mechanism with release mechanisms such as Fickian diffusion, Non-Fickian transport, Case II transport, Super case II transport [24, 29]. The release kinetics is used to predict the release before the drug was conducted [30]. The release kinetics of red ginger oleoresin from microcapsules with different wall materials were determined using equations 4–8. Table 1 shows the constants obtained from various release kinetics models. Table 1 shows the release kinetics constant obtained from the R² value from various models, based on the differences of wall material used. The R² value indicates the suitability fitting between each model's experimental and calculation data. The R² values obtained from equations 4 to 8 are between 0.97 – 0.99. The best model for red ginger oleoresin release from all types of wall material used is Korsmeyer Peppas and Peppas – Shalin model. Fig 4 shows a good fitting between experimental and calculated data from red ginger oleoresin microcapsules using 2% (w/v) maltodextrin as wall material.

![Figure 4. Cumulative release red ginger oleoresins from microcapsules using maltodextrin 2% (w/v) as wall material, ratio between experimental and calculated data from release kinetics model: [A] zero order, [B] first order, [C] Higuchi model, [D] Korsmeyer-Peppas model, [E] Peppas-Shalin model.](image)

Figure 4 shows a good fitting between experimental and calculated data for all release kinetics models. The Korsmeyer Peppas model was used to determine the release mechanism from the value of n. Table 1 shows the n value of microcapsules using carrageenan, maltodextrin, and their mixture at 1:1 and 2:1 weight ratio is greater than 0.43. Based on Ritger and Peppas [30], the release mechanism at range 0.43 < n < 0.85 is an anomalous (non-Fickian) transport, which indicates that the release mechanism was controlled by Fickian diffusion and swelling transport. Red ginger oleoresin microcapsules with combined maltodextrin-carrageenan as wall material at weight ratio 1:2 showed a value of n < 0.43. This condition indicates that the release mechanism occurs by diffusion. The nature of carrageenan, which is difficult to dissolve in water, makes the red ginger oleoresin release mechanism occur by Fickian diffusion.

4. Conclusion

Red ginger oleoresin microcapsules were successfully prepared by spray drying using two different wall materials and their mixture with different weight ratios. The type of wall material determines the encapsulation efficiency and the amount of red ginger oleoresin released from the microcapsules. The highest encapsulation efficiency was obtained from microcapsules with combined maltodextrin - carrageenan wall materials at ratio 1:2, and the lowest encapsulation efficiency resulted from microcapsules with combined maltodextrin and carrageenan wall materials at ratio 1: 1. The addition of carrageenan formed a more rigid microcapsule wall characterized by low cumulative release and high encapsulation efficiency. Release kinetics of red ginger oleoresin with the good fitting of all microcapsules wall materials were Korsmeyer-Peppas and Peppas-Shalin models with R² values 0.97 – 0.99. The release mechanism based on the value of n is anomalous (non-Fickian) transport and Fickian diffusion.

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