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Formulation and Physical Properties of Citronella Oil Emulsion on Differences in Emulsifiers With The Addition of Maltodextrin

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ABSTRACT

Various studies have shown that citronella oil is a fatty oil that is efficacious for reducing microbial activity in food. Emulsions are preparations containing two immiscible substances, usually water and oil, where the liquid which when dispersed becomes small particles in another liquid. The combination of maltodextrin and citronella oil requires an emulsifying agent. Methyl cellulose and gum arabic have been widely used with maltodextrin because they can increase the stability of the emulsion formed. Apart from being an emulsifier, the addition of methyl cellulose and gum arabic aims to strengthen the resulting coating layer. This study using response surface methodology (RSM) for the formulation of thin layer emulsion-based edible coating materials, studies the characteristics and stability of the thin layer emulsion system on edible coating materials, and studies the effect of storage temperature on the stability of the thin layer emulsion system on edible coating materials by observing the physical properties of the emulsion which includes the type emulsion, droplet size, and creaming index on sedimentation volume for 1 month of storage on variations of emulsifier methyl cellulose and Arabic gum and addition of maltodextrin. The results of observations based on tests on the type of emulsion, droplet size, and creaming index on the sedimentation volume for 1 month of storage on variations in emulsifier methyl cellulose and Arabic gum and the addition of maltodextrin showed that there was a significant difference. © 2023 The Authors. Published by Elsevier Ltd. This is an open access article under the CC BY-NC-ND

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1. Introduction

Emulsions are one of which is in the fields of food, chemistry, and pharmacy. According to the continuous phase difference, it can be grouped into Oil in Water (O/W) emulsion, Water in Oil (W/O) emulsion and multiple emulsion [1]. In recent years, there has been a strong interest in the food industry in exploiting the potential of citronella oil as an edible coating in functional foods and supplement products due to its potential health benefits and longevity [2]. The process of making edible coatings made from citronella oil utilizes the emulsification process, emulsifying oil in water (O/W). An O/W emulsion is a thermodynamically unstable system involving two immiscible liquids (generally such as water and oil) on the principle that the oil is distributed in the water [2,3].Table 1.

Emulsion stability is determined by process, temperature, pH, type and concentration of emulsifier. As a result, thermodynamic equilibrium is attained through a number of mechanisms, including Ostwald maturation, flocculation, coalescence and creaming or sedimentation [1]. Emulsion stability is a determining factor in determining the shelf life of food products [3]. The preparation method is a factor for the stability of the o/w emulsion. Some of the methods used to make emulsions are ultrasound (US), mechanical treatment, and high-speed mixing in a homogenizer [3]. The food industry often uses the principle of homogenization with stirring speed and high pressure because it produces high efficiency [4].

Citronella oil can reduce microbial activity and maintain the physical quality of bell pepper because it contains phenolic components and flavonoids which are antioxidants [5]. Due to the hydrophilic nature of maltodextrin, the combination of maltodestrin and

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Table 1

Emulsification Formulation

Composition	[a]	[b]	[c]
Maltodextrin DE 10	10%	20%	30%
Oil (Citronella Oil)	6%	6%	6%
Emulsifier	10%	10%	10%
Coating Agent (Shellac)	5%	5%	5%
Water	79%	59%	49%

citronella oil requires an emulsifying agent. Methyl cellulose is widely used with maltodextrin because it can increase the stability of the emulsion formed [6,7]. Apart from being an emulsifier, the addition of methyl cellulose aims to strengthen the resulting coating layer. This is due to the molecular weight of maltodextrin which is an intermediate so that the resulting coating is weak [8]. Therefore, additional material is needed that can strengthen the resulting coating layer.

The main objective of this study was to determine the maltodextrin based citronella oil emulsion formulation with methyl cellulose and arabic gum emulsification as emulsifiers on the stability of 6% o/w coconut oil emulsion. The selection of maltodextrin-based emulsifiers with methyl cellulose and arabic gum is in high demand because label-friendly products contain natural and sustainable ingredients. To find out the optimal formulation using RSM and the emulsion stability approach by analyzing the creaming index and droplet size of 6% citronella oil in a water emulsion made with high mixing speed in a homogenizer.

2. Materials and methods

2.1. Materials

The materials used in this study were methyl cellulose as an emulsifier, maltodextrin DE 10 as an emulsion stability enhancer, citronella oil and water, shellac as a coating agent purchased from Sigma-Aldrich Chemical Reagent Co., Ltd. All other chemicals used were analytical grade and purchased from Indrasari chemist shop. While the main tools used are homogenizer IKA T-10 ULTRA TUR-RAX, UV–VIS spectrophotometry, binocular microscope with 10x magnification, and laboratory analysis tools.

2.2. Emulsification preparation

An O/W emulsion with citronella oil and water was obtained by high pressure homogenization. The surfactant system and surfactant concentration used in this article are methyl cellulose as an emulsifier, maltodextrin DE 10 as an emulsion stability enhancer, citronella oil and water, shellac as a coating agent. 5% w/w shellac was diluted in 500 ml of distilled water and then heated at 40 $^\circ C$ – 45 °C. Furthermore, aqueous phase surfactant solution was prepared by dispersing 6% w/w citronella oil, 10% w/w, 20% w/w, 30% w/w maltodextrin DE 10 and 10% w/w methyl cellulose into water 49% w/w, 59% w/w, 79% w/w and stir the mixture of ingredients with homogenizer for at least 10 min to ensure complete dissolution. The last step is to add the coating agent, namely shellac 5% w/w, into the homogenized chili mixture. A coarse emulsion is obtained by mixing the oil phase and the water phase, using an IKA T100 homogenizer stirrer at 4000 rpm for 10 min at incubator. For several experiments, repeat each formulation. After preparation, all emulsions were stored in an incubator at temperatures of 30 °C, 70 °C, and 110 °C. check every 0 days, 1 day, 3 days, 5 days, 10 days, 15 days, 20 days, 25 days, and 30 days for 1 month.

2.3. Characterization of Maltodextrin-Based citronella oil emulsion formulation on methyl cellulose emulsifier

• Measurement of Emulsion Stability by Determining Creaming Index

Measurement of emulsion stability by calculating the creaming index using a UV–Vis spectrophotometer with a wave size of 500 nm.

The emulsion prepared by AS treatment or homogenization of stirring speed and high pressure was diluted 500 times with 0.05% emulsion yield and the absorbance (A) of the diluted emulsion was measured using a UV–VIS spectrophotometer at a wavelength of 500 nm. Emulsification activity index (EAI, m2/g) was calculated according to the following formula:

$EAI = \frac{2303x2xAxN}{Cx \varnothing x10000}$

Information:

A = Absorbance at 500 nm:

- B = Emulsion diluent factor N (10).
- C = Emulsifier concentration (10%)

 $\emptyset = Oilvolume fraction(6\%)$

• Measurement of Droplet Size with a 10x Magnification Microscope

Droplet size analysis using a binocular microscope with oil sample preparation of about 0.01 ml of stabilized emulsion sample before and after treatment, then, place it on the slide and cover with a cover slip after 30 min of equilibrium, the sample is examined at room temperature with 10X magnification at under a binocular microscope. Cahaya is equipped with a digital camera that is connected to the Optical Application on a Computer. Then analyzed using the Image J software application to determine droplet size.

• Statistic Analysis

Statistical analysis was performed using Origin V9.0 software. Calculation of Creaming Index and droplet size analysis measurements was carried out with three repetitions and the results obtained were average ± standard deviation (SD). RSM analysis was performed to determine the most optimal formulation.

3. Experimental design and Statistical analysis

In this experiment using the RSM method based on central composite design (CCD) to obtain the most optimal Emulsion Formulation in the manufacture of edible coatings. At 3 levels of independent variables ranging from 1 to + 1, 18 experiments were produced with three independent variables namely, Maltodextrin (A); Air (B); and incubator temperature (C).

Incubator temperature: 30 °C, 70 °C, and 110 °C, within 1 month. By comparison the emulsifier Methyl Cellulose and Arabic Gum.

4. Results and discussion

4.1. Effect of maltodextrin concentration on citronella oil emulsion results

The effect of maltodextrin concentration and the ratio of citronella oil to maltodextrin on the yield of citronella oil microcapsules is shows in Fig. 1. In general, it can be seen that the greater the con-



Fig. 1. Results of the Effect of Maltodextrin Concentration on Emulsion Results.

centration of maltodextrin and the ratio of citronella oil to maltodextrin used, the greater the yield of citronella oil microcapsules product obtained. It can be concluded that the concentration of maltodextrin and the ratio of citronella oil to maltodextrin have an effect on increasing the yield of citronella oil microcapsules.

The stability of the active ingredients during the storage process is closely related to the amount of oil on the surface of the microcapsules. The presence of oil on the surface of the microcapsule wall is not desired. This is because the oil is not encapsulated, namely the oil that is on the surface. will be exposed to the surrounding environment and can be damaged [3]. The functional characteristics of maltodextrin are related to the value of DE and empirically become a guideline for determining its application. For example, maltodextrin with DE10 is commonly used for encapsulating aromas, flavors, and food products. Variations in DE values produce maltodextrin with different physico-chemical properties, such as viscosity, density, hygroscopicity, osmolality, and compactness. It can be concluded that the addition of DE 10 maltodextrin by 30% increased the ability to form films for the manufacture of edible coatings.

4.2. Creaming index results on droplet size

The Result shows in Fig. 2 using microscope with the ImageJ application to determine the size of the droplets in the mixture using methyl cellulose and arabic gum emulsifiers. The hydrophilic nature of maltodextrin, the combination of maltodextrin and citronella oil requires an emulsifying agent. Therefore, methyl cellulose and Arabic gum are good emulsifiers in the market. The difference in this emulsifier lies in the level to strengthen the resulting coating layer and is closely related to the droplet size. The smaller the droplet size, the more stable it is and the smaller the speed of creaming, so that the separation that occurs is not large.

Therefore, the greater the creaming index, the less separation occurs. Moisture content is one of the important parameters related to product stability during the storage process and ease of flow of microcapsules products [1]. The higher the mixing temperature, the more easily the droplets combine so that the droplets have a larger size. The longer the storage time at 30 °C, the easier it will be to form droplet sizes so that the droplet sizes become larger. In Arabic gum has a smaller droplet size 45.358 μ m while in methyl cellulose it has an average droplet size of 69.424 μ m. When compared to emulsifiers, the emulsion using Arabic gum was easier to mix and stable.

4.3. Result of relationship of emulsion stability with creaming index level

RSM is used to determine the effect of temperature on emulsion stability and the temperature with the most optimal droplet size. Eighteen experiments were carried out randomly. Fig. 3 shows the results of the relationship between emulsion stability and creaming index for each point based on the CCD experimental plan. The influence of process variables on the emulsion and droplet size by describing the three-dimensional surface curve to the three independent variables whose EAI remains at the control level. The 3D curve of the fixed variable response is shown in Fig. 3 (a,b).

Creaming occurs when the emulsion droplets are moved to the top or bottom according to the difference in density between the dispersed phase and the continuous phase. The degree of creaming is affected by the droplet size, the viscosity of the dispersed phase, and so on. However, the state of the emulsion changes over time. Also, the crumbling process changes the quality of the emulsion product [1]. Homogenization with high agitation speed is suitable for liquid products because it is a continuous non-thermal process. In this process, the mixed material is subjected to a high mixing speed which will result in high pressure through the narrow homogenization gap. The shear strength of the homogenization process at high agitation speeds and pressures allows the mixed molecules to coalesce into large mixed molecules. Therefore, homogenization with high stirring speed and pressure is often used to produce o/w emulsions [2]. Fig. 3(a) illustrates the effect of temperature with the stability emulsion on the first day. The results showed that the emulsion increased with increasing droplet size in the emulsion. Fig. 3 (b) shows the effect of storage temperature on droplet size on the third day which shows that droplet



Fig. 2. Droplet Size Results (a) Methyl Cellulose; (b) Arabic Gum.



Fig. 3. Response surface plots describe the effects of (a) temperature on emulsion stability; (b) temperature with droplet size.

size increases with increasing storage temperature and storage time. Emulsion stability slightly increases with droplet size.

5. Conclusion

The results of research on making emulsions for edible coating products show that the concentration of maltodextrin and the ratio of citronella oil greatly affect the emulsion product. The citronella oil microcapsule product with the highest yield can be identified through the results of the creaming index in each mixture so that it can be concluded that the results in the mixed treatment with a concentration of 30% maltodextrin coating material and a ratio of citronella oil to maltodextrin 1:5 produces the highest yield in the creaming calculation results index. RSM was used to study the emulsion formulations by emulsification process using a homogenizer. Homogenization process with high agitation rate for the manufacture of oil-in-water emulsions by carrying out several analyzes related to emulsion preparation and droplet size. These results indicate that the process of homogenizing citronella oil in air with modifications to the use of maltodextrin was able to increase film formation for the manufacture of edible coatings.

Data availability

The data that has been used is confidential.

Declaration of Competing Interest

The authors declare that they have no known competing financial interests or personal relationships that could have appeared to influence the work reported in this paper.

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