Mechanical Properties of Whey Composite Edible Film with the Addition of Clove Essential Oil and Different Types of Plasticizer

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Abstract. The purpose of this study was to determine the mechanical properties of the composite whey edible film with the addition of clove oil and different types of plasticizers. This research was conducted as a laboratory experiment and the variables observed were the mechanical properties of the composite whey edible film. This research was conducted using a Factorial Completely Randomized Design consisting of Factor A (clove oil concentration: 5%; 10% and 15%) and Factor B (a type of plasticizer): sorbitol and polyethylene glycol. The results showed that the percentage value of elongation with the use of clove oil was 46.00-63.01% and the use of plasticizers resulted in 49.56-57.33%. The value of tensile strength using clove oil resulted in 7.76-7.9 N and the use of plasticizers from 7.74-8.05N. WVTR film with the use of clove oil 7,00-7,31 g/m2.day-1 and the use of plasticizer resulted in a value of 7,09-7,16 g/m2.day-1, for the microstructures showed a film surface that still showed oil droplets and no homogenization process but no cracks occurred in the film. Composite whey edible film with the addition of clove oil and a different type of plasticizer can produce good film mechanical properties.

1. Introduction

Environmentally friendly technology aimed at obtaining food safety, quality and easy handling properties has received great attention for edible film research, which can later be applied to food products. Edible film is defined as a thin edible film that is formed on the surface of food or food products as the main packaging which has various benefits. The advantages of edible film itself are that it has biodegradable properties, can protect packaged products as well as a carrier system for active substances such as antioxidants, antimicrobials, giving taste and color.

Edible film requires the best formulation to produce the best film structure, such as with the addition of protein and polysaccharides. Protein-based edible films have interesting properties, whey protein has special properties that make them very suitable for packaging for food products. Its excellent nutritional value and various functional properties such as solubility in water and the ability
to act as an emulsifier are important factors for the formation of edible films. Among the types of protein, whey is one of the most preferred packaging materials (Schimd et al., 2012).

The use of protein and polysaccharides will form edible films which have good mechanical properties, but are bad in terms of moisture, this is due to their hydrophilic properties. The use of oil can be added to the edible film solution to obtain an emulsified film which only requires a drying process (Ma et al., 2014), the addition of essential oils is a very attractive representation of film formulations with the aim of making them natural packaging (Sánchez-González et al., 2011; Silva-Weis, 2103) Generally, essential oils act as a good moisture barrier (Hambleton et al., 2012). Therefore, the lipid-composite whey edible film can have a good moisture holding capacity. The addition of plasticizers is also very much needed to get a film that has a level of elasticity so that the film is not easily broken. Plasticizers from the group commonly used to be mixed into film solutions such as sorbitol, glycerol and polyethylene glycol. Baldwin et al., 2012 stated that plasticizers have the ability to intermolecular forces and will increase the flexibility of the edible film by widening the molecular free space.

Thus, the main objective of this research is to optimize the properties of edible whey film composites through the addition of clove oil and a type of plasticizer to assess mechanical properties (tensile strength, elongation, water vapor transmission rate, cross-sectional structure by scanning electron microscopy).

2. Materials And Methods

The materials used in this research are whey powder, konjac powder, sorbitol plasticizer, polyethylene glycol plasticizer, distilled water, wrapping, label paper, silica gel, plastic, aluminum foil, alcohol 70%, rolled tissue, water, clove essential oil.

The tools used in this study were petri dish, micropipette, thermometer, Erlenmeyer, magnetic stirrer, hot plate stirrer, petri dish, freezer, micrometer screw, measuring cup, measuring tube, digital HF 500 gauge, desiccator, water pass, pH meter, stopwatch, scissors, ruler, digital scale, a set of test equipment (tensile strength and elongation), Scanning Electron Microscope, desiccator, knife, clamp, and others.

The research was conducted in a completely randomized design with five replications. All of the data were analyzed by using analysis of variance (ANOVA) and followed with the least significant difference (LSD) test at P<0.05 level to determine significant differences and P<0.01 for highly significant differences. The analysis was employed in SPSS 16.0 program.

1. Edible Film Preparation

Two grams of 8% (w/v) whey powder was mixed with 0.5 g (w/v) konjac, clove oil 5%; 10%; 15% (w/v) and diluted with distilled water at 25 mL. The solution was then added with glycerol as much as 30% of the whey and konjac solution according to the treatment. The solution was then heated at 90°C ± 2°C in the hot plate and simultaneously mixed by using a magnetic stirrer at 250 rpm for 30 m. The film solution was then poured in the petri dish and dried at room temperature for 24 h. The edible film was then packaged by using a paper wrap for 2 d before analyzed (Fahrullah et al., 2020b).

2. Elongation at break.

The elongation at break was measured by using Universal Instrument Tensile Strength Meter (ASTM D882-1). The film was cut at 10 x 5 cm area and stretched at 50 mm/ minute speed. The elongation was then measured by following formula:

Elongation (%) = L/L0 x 100%.

Description:
L  = length of the edible film at break (mm)
L0  = initial length (mm)
3. Tensile strength.

The edible film was cut at 8 x 3 cm with the diameter at 1.5 cm and hooked horizontally on a clamp on both long sides. The maximum tensile strength was measured when the film showed the sign of breakage during the pullout (Wittaya. 2014).

4. Water vapor transmission rate.

The water vapor transmission rate (WVTR) was observed by firstly cut the edible film into circle with 2.8 cm diameter. The film was then placed to cover a glass filled with 3 g silica gel. The covered glass was then placed into desiccator. The sample weight was measured every 24 hours for 5 days. The WVTR is presented in the unit of g.mm\(^{-2}\).day\(^{-1}\), and calculated by following formula:

\[ \text{WVTR} = \frac{n}{(t \times A)} \]

Description:

\[ n = \text{Weight change (gram)} \]
\[ t = \text{time (day)} \]
\[ A = \text{Edible film surface area (mm}^2\) \]

5. Edible film microstructure.

The film microstructure was observed with scanning electron microscopy (SEM JEOL JSM 5310 LV). The film was cut at 0.5 x 0.5 cm area, then placed on carbon covered plate and coated with gold. The sample was then placed on the SEM device for microstructure observation.

3. Results And Discussion

Elongation

The elongation of the resulting whey composite edible film ranged from 40.66 to 66.67% (Table 1) where the treatment of using 15% clove oil with the use of sorbitol plasticizer produced the highest value. This elongation value has not met the edible film standard, which is at least 70% (Japanese Industrial Standard, 1975). The results of this study were higher than (Fahrullah, 2020a) with the elongation of using sorbitol plasticizer only 29.86% and the use of polyethylene glycol produced 28.66%.

| Clove Oil (%) | Plasticizer          | Average          |
|--------------|----------------------|------------------|
|              | Sorbitol             | Polyethylene Glycol |               |
| 5            | 51.33 ± 6,49         | 40,66 ± 6,83      | 46,00\(^a\)    |
| 10           | 54,00 ± 14,98        | 48,67 ± 10,43     | 51,34\(^a\)    |
| 15           | 66,67 ± 14,33        | 59,34 ± 10,11     | 63,01\(^b\)    |
| Average      | 57.33                | 49.56             |                 |

\(^a\) Superscripts in the same column show very significant differences (P < 0.01)

Analysis of variance showed that the different clove oil concentration treatments showed a very significant difference (P < 0.01), while the use of plasticizers did not provide significant differences (P > 0.05). The higher the addition of ± clove oil, the lower the elongation value of both sorbitol and polyethylene glycol. This effect can be attributed to the development of discontinuities in the polymer network resulting from the addition of clove oil. In addition, this increase is associated with the effect of the plasticizer that enters the film matrix, because the use of this plasticizer has an effect on increasing the elasticity of the film. The increase in the elongation of composite whey edible film also occurred in the addition of olive oil (Ma et al., 2014; Pereda et al., 2012).
Tensile Strength
The tensile strength of the resulting composite whey edible film ranged from 6.82-8.94 N, where the treatment with 5% clove oil and polyethylene glycol plasticizer produced the highest tensile strength value. The results of this study are not much different from Fahrullah's (2020) research which resulted in a tensile strength value using sorbitol of 7.82 N and polyethylene glycol of 8.20 N.

Table 2. Tensile strength of the composite whey edible film with the addition of clove oil and plasticizer

| Clove Oil (%) | Plasticizer | Sorbitol | Polyethylene Glycol | Average |
|---------------|-------------|----------|---------------------|---------|
| 5             |             | 6.86 ± 0.83 | 8.94 ± 1.54        | 7.9     |
| 10            |             | 6.82 ± 1.56 | 8.3 ± 0.75         | 7.56    |
| 15            |             | 7.66 ± 0.95 | 7.86 ± 0.33        | 7.76    |
| Average       |             | 7.74\textsuperscript{a} | 8.05\textsuperscript{b} |         |

\textsuperscript{ab} Superscripts on the same line show very significant differences (P <0.01)

Analysis of variance showed that the use of clove oil did not provide a significant difference (P> 0.05) while the use of plasticizers provided a significant difference (P <0.01) between treatments. General the tensile strength depends on the concentration of the film formula and the conditions of preparation before the film is formed (Siracusa et al., 2018). The strong interaction between polymer and plasticizer provides a cross-linking effect which will reduce the free volume and mobility of polymer molecules. The incorporation of clove oil into whey edible film will affect the strength of the film produced (Soazo et al., 2011). The difference in tensile strength values depends on the type of oil used as well as the composition of the film. Films with high tensile strength generally have a lower elongation percentage so it is necessary to measure elongation and tensile strength simultaneously (Galus and Renart, 2013).

WVTR (Water Vapor Transmission Rate)
WVTR value whey edible film composite yield between 6.82-7.47 g/m$^2$.day$^{-1}$, where the addition of clove oil treatment 15% with plasticizer sorbitol produces the lowest values. This value is still not meet the standards of JIS (Japanese Industrial Standard, 1975) which generates a maximum value of 10 g/m$^2$.day$^{-1}$

Table 3. WVTR composite whey edible film with the addition of clove oil and plasticizer

| Clove Oil (%) | Plasticizer | Sorbitol | Polyeten Glikol | Average |
|---------------|-------------|----------|-----------------|---------|
| 5             |             | 7.47 ± 1.05 | 7.15 ± 0.67     | 7.31    |
| 10            |             | 6.99 ± 0.92 | 7.15 ± 0.67     | 7.07    |
| 15            |             | 6.82 ± 0.44 | 7.17 ± 0.71     | 7.00    |
| Average       |             | 7.09      | 7.16            |         |

Analysis of variance showed that the addition of clove oil and the use of plasticizer gave a significant difference (P> 0.05) to the WVTR value of composite whey film edible. Table 3 shows that the addition of clove oil which increases will result in a decrease in the WVTR value of the composite whey edible film. This is because clove oil has hydrophobic properties and the combination of whey-konjac-clove oil has an effect on increasing hydrophobic properties. This will have an impact on the film which is increasingly difficult for moisture to penetrate. Films containing plasticizers will create a plasticizing effect so that they can interact and reduce the cohesiveness of the whey matrix, which will increase solubility in water, but on the other hand, the use of oil can reduce solubility in water (Galus and Kadzinska, 2016). In addition, it is also influenced by the number of polar groups contained in the
polymer so that it will affect the diffusivity and solubility of water molecules in the film matrix (Kokoszka et al., 2010).

**Edile Film Microstructure**

The microstructure of edible film was tested using a scanning electron microscope (SEM) with a magnification of 1000x to evaluate the homogeneity of its surface and structure. Specifically for this treatment, microstructural testing is only carried out on the best treatment seen in the type of plasticizer, where the plasticizer sorbitol is the best compared to polyethylene glycol in terms of the elongation percentage produced. Figure 1 shows the surface structure of the composite whey edible film. Figure 1 shows the surface structure of the composite whey edible film.

![Figure 1. Composite whey edible film microstructure with the addition of clove oil (a) 5%; (b) 10%; (c) 15% with sorbitol plasticizer.](image)

The resulting composite whey edible film showed elastic properties and the surface of the structure had no cracks. Figure 1 shows an inhomogeneous surface, because the matrix of the whey protein seems to be disturbed by the presence of clove oil, resulting in an emulsion process. The addition of clove oil is both 5%, 10% and 15%. Most of the oil droplets are visible on the surface of the film, the more clove oil is added, the more oil droplets on the surface are. The aggregation of oil droplets during the film drying process is clearly seen in Figure 1. The phenomenon of aggregation and coalescing begins after homogenization without emulsifier and occurs during drying. The formation of oil droplets during drying can cause disturbance in the film matrix.

Whey protein plays an important role in stabilizing the emulsion by forming a stabilizing layer around the oil droplets. This shows that whey protein has the ability to form a barrier around oil droplets during the migration process. But on the other hand, the presence of clove oil in this film can increase moisture absorption and water vapor mobility, therefore whey composite edible films made
from hydrocolloid and lipid materials have an advantage related to the migration of water molecules or active compounds that are good for coating materials on food products.

4. Conclusion

Composite whey edible film with the addition of clove oil and a different type of plasticizer can produce good film mechanical properties. The addition of clove oil can affect the resulting changes in the structure of the film and can reduce the humidity which will be very important in the application of films with high moisture content products. The edible film produced can be considered as a natural packaging material that has the potential to increase the shelf life of a product.

Acknowledgements

The authors would like to thank the Indonesian LPDP (Educational Fund Management Institute) for funding this research.

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