ALCHEMY Jurnal Penelitian Kimia
Official Website: https://jurnal.uns.ac.id/alchemy
ISSN 1412-4092 EISSN 2443-4183

Development of Bioplastic from κ-Carrageenan and β-Cyclodextrin

Nurlaida Nurlaida*, Maipha Deapati Arief, Neny Rasnyanti M. Aras, Nurhajawarsi Nurhajawarsi

Department of Analytical Chemistry, Akademi Komunitas Industri Manufaktur Bantaeng
Nipa-Nipa, Pakjukukang, Bantaeng, South Sulawesi 92461, Indonesia

Corresponding author: nurlaida@akom-bantaeng.ac.id

DOI: 10.20961/alchemy.22.2.118703.434-441

Received 12 May 2026, Revised 14 August 2026, Accepted 18 August 2026, Published 30 September 2026

Keywords:

biodegradable plastic;
biopolymer;
β-Cyclodextrin;
κ-Carrageenan.

ABSTRACT. κ-Carrageenan is a type of polysaccharide that has properties such as thickening and gelling ability. These attributes contribute greatly for its application in the development of biodegradable plastic. However, it still suffers from poor mechanical properties and solubility. β-Cyclodextrin is an oligomer that due to its structure, acts as a cage for substances with similar polarity. This study aims to develop a bioplastic with combination of κ-Carrageenan and β-cyclodextrin, using the latter as a shield to protect κ-Carrageenan and to enhance their mechanical properties. The concentration of β-Cyclodextrin was varied as kb-0; kb-10; kb-20 which the numbers represent the ratio of β-Cyclodextrin to κ-Carrageenan, while concentration of κ-Carrageenan remained constant. Incorporating β-Cyclodextrin into the film matrix gave results of thicker films, higher water vapor permeability, as well as tensile strength. The opposite effects observed in lower value of water content, swelling capacity, and elongation at break. At first, thread-like surface was detected when small β-Cyclodextrin was added, and become more granule as its level increased.

INTRODUCTION

The majority of food packaging comes from petroleum-based plastic, which is a non-renewable type of packaging. Polyethylene, polypropylene, and polyethylene terephthalate are plastics that are widely used for this purpose. One of their properties is that they are durable, have a good thermal and mechanical properties, as well as its processing ability, hence, this leads to a condition where the accumulation of these plastics is rapidly increasing over time. An extremely huge amount of plastic ends up in open water and landfills every year, making them as threat to the environment, including aquatic species. Another drawback of conventional plastic is microplastics, which is formed when the plastic is decayed, and it can cause health problems if it is absorbed by organisms due to its toxicity (Barizão *et al.*, 2020; Carina *et al.*, 2021; Bidari *et al.*, 2023).

Extensive efforts in developing polymers, natural or synthetic, are being done to minimize the dependence on plastics. Biodegradable film, made from polymers, which consist of thin layers of materials that function as packaging to protect the food inside by preventing moisture, gases, and microbial contamination in contact with the food. An alternative to these plastics, where the properties are expected to be similar to plastics, yet biodegradable, is now more necessary than ever. Terms like smart packaging also emerge as a more advanced attempt to replace plastic. Smart packaging often contains antibacterial, antioxidant, and other properties that offer a promising alternative to conventional plastic (Bidari *et al.*, 2023; Dmitrenko *et al.*, 2025). This study aims to develop a food packaging that is made from natural materials and easy to degrade.

The advancement of technology in packaging has opened the possibility to produce films that come from renewable materials. These materials, usually polymers, are often biodegradable and environmentally friendly and are being moved toward substitution for plastics. Different types of polymers have been proposed for this purpose, for instance, polysaccharides (starch, chitosan, cellulose, gelatin, waxes, etc.), lipids, and proteins (soy protein isolate). There are also other types of polymers, such as synthetic and microbial polymers, that have been studied for the same objective as well (Ruiz-Navajas *et al.*, 2013; Fonseca-García *et al.*, 2021; Wen *et al.*, 2021; Díaz-

Cite this as: Nurlaida, N., Arief, M. D., Aras, N. R. M., and Nurhajawarsi, N. (2026). Development of Bioplastic from κ-Carrageenan and β-Cyclodextrin. *ALCHEMY Jurnal Penelitian Kimia*, 22(2), 434-441. doi: https://dx.doi.org/10.20961/alchemy.22.2.118703.434-441.

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Montes, 2022; Dirpan *et al.*, 2023; Tafa *et al.*, 2023; Tian *et al.*, 2024). Among all those natural polymers, polysaccharides are probably the most studied as one of the ingredients to develop environmentally safe packaging. The characteristics of polysaccharides, such as biodegradability, biocompatibility, and non-toxicity, are what make them stand out (Liu *et al.*, 2019; Nešić *et al.*, 2019).

Carrageenan is a gel-forming, linear sulfated polysaccharide that is abundantly found as the main component of red seaweed. The structure of carrageenan alternates between 3-linked β-D-galactopyranose and 4-linked α-galactopyranose or 3, 6-anhydro-α- galactopyranose. There are three types of carrageenan based on the amount and position of the ester sulphate groups and the number of 3, 6-anhydro-galactose forming the carrageenan. Those types are κ-, ι-, and λ-carrageenan (Campo *et al.*, 2009; Necas and Bartosikova, 2013; Liu *et al.*, 2019). The exceptional thickening, gelling, and stabilizing ability of carrageenan is what makes it stand out among other types. Besides packaging, carrageenan is also famous in the pharmaceutical industry (as a component for bone regeneration and capsule shell), cosmetic, and textile industry. Carrageenan also has high robustness, fair compatibility, and viscoelasticity which resulting in its utilization in other areas (Campo *et al.*, 2009; Supraja *et al.*, 2025; Soraya *et al.*, 2025). A lot of reports show that when mixed with other substances, film made from carrageenan improves its physical properties. A combination of starch and carrageenan created a film with better tensile strength (Shahbazi *et al.*, 2018; Dmitrenko *et al.*, 2025). Another study reported that adding nanocellulose resulted in a film with enhanced water resistance, hydrophobicity, and mechanical strength, as well as reduced water vapor permeability (Dmitrenko *et al.*, 2025). Gel strength is reported to increase with the presence of carrageenan in gelatin capsule waste (GCW) (Sanprasert *et al.*, 2025).

Cyclodextrin is an enzyme-modified starch derivative that is being used in pharmaceutical and food industry for decades. It consists of glucopyranose building units that are linked with α-1,4 glycosidic bonds. Cyclodextrin has a hydrophilic surface due to its hydroxyl groups, while inside it has lipophilic property. There are three kinds of cyclodextrin: α-cyclodextrin (ACD) formed by six glucopyranose units, β-Cyclodextrin (BCD) with seven units, and γ-cyclodextrin (GCD) that has eight units of glucopyranose (Szente and Fenyvesi, 2017). Cyclodextrin is a cyclic glucopyranose oligomer that acts like a cage, and forms a truncated cone-shaped lipophilic cavity. Its structure makes it possible for it to enclose substances that possess similar polarity with its cavity. The encapsulation and complexing ability of cyclodextrin are the main reason it is widely used in medical and pharmaceutical industry, because it offers a solution for poor solubility, stability, and dissolution cases of some materials and can improve delivery, lessen toxicity, and enhance intake of some medicine (Elewi *et al.*, 2025; Karali *et al.*, 2025; Márton *et al.*, 2025).

Cyclodextrin has been shown to possess antifungal activity of *C. albicans* while also have the ability to reduce biofilm formation, depends on concentration of it (Elewi *et al.*, 2025). Márton *et al.* (2025) revealed that using cyclodextrin and mixture of κ-carrageenan and gelatin, to form nanofibers, as a capsulation metdhod for quercetin had improved its physical properties. The size of the nanofiber is reduced while enhance the tensile strength and modulus, as well as improve the thermal stability of the fiber. By adding cyclodextrin into alginate hydrogel solution, the hydrogel beads increses its water absorption, possibly by enhancing its pores (Karali *et al.*, 2025). Magnoliae essential oil (MEO) and β-Cyclodextrin (β-CD) (SA/MEO@β-CD coating) exhibits effects of reducing lipid oxidation and preventing microbial activity in order to prolong the shelf life of Kyoho grapes. It opens an opportunity to create a packaging for short-life foods (Tian *et al.*, 2024).

These studied have clearly established the advantages of carrageenan and cyclodextrine for their respective functionalities. It is proposed that the presence of cyclodextrine will further enhance the characteristics of biodegradable plastics made from carrageenan, whose drawbacks come from their solubility and mechanical properties. This study aims to combine κ-carrageenan and β-Cyclodextrin (β-CD) and search the right formula to produce bioplastic that will reduce our dependency for conventional plastic, which based on the authors' knowledge, have never been studied before this paper.

RESEARCH METHODS

Analytical balance, Scanning Electron Microscope (JOEL JCM 6000plus, Laboratorium Mikrostruktur Universitas Muslim Indonesia), digital micrometer (Mitutoyo), Texture analyzer (Shimadzu, Laboratorium Metalurgi Fisik Universitas Hasanuddin), FTIR IRPrestige-21 (Shimadzu, Laboratorium Kimia Terpadu Universitas Hasanuddin), polystyrene petri dish (50 mm), hygrometer, desiccator, thermometer, dehumidifier (Blue Sky), oven, centrifugal tube 50 mL (Eppendorf), and common glassware. The ingredients used in this

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research are κ-Carragenan (Indonesia Handmade), β-Cyclodextrin (β-CD), glycerol p.a., ethanol 70%, silica gel, and aquadest.

Preparation of the κ-Carragenan/ β-Cyclodextrin (β-CD) Film

The bioplastics were prepared according to Liu *et al.* (2019) and Panrong *et al.* (2019) with some modifications. 150 mL of aquadest was heated to reach 90 °C. To the solution, κ-Carragernan, β-Cyclodextrin (β-CD), and glycerol p.a. were added in the amounts as described in Table 1. The mixed solution stayed at this temperature for about 30 minutes until all the materials dissolved completely. 40 mL of the solution was poured onto polystyrene petri dish and dried at 60 °C until a dry film sheet was formed. If it is not used, the film was stored at room temperature.

Table 1. The formula between κ-Carragernan, β-Cyclodextrin (β-CD), and glycerol p.a. (w/w).

Sample code	κ-Carragernan (gram)	β-Cyclodextrin (β-CD) (% to κ-Carragernan)	Glycerol p.a. (% to κ-Carragernan)
kb-0	1.6	0	25
kb-10	1.6	10	25
kb-20	1.6	20	25

Characterization of Bioplastic Film

Thickness

The thickness of the film was measured using a digital micrometer with an accuracy of 0.001 mm. The measurement was done at four different spots on the film. Each measurement was done three times.

Water Content and Swelling Test

Water content was measured by drying the film at 105 °C until the weight was constant. The formula used can be seen as follows in Equation 1.

$$Water\ content\ (\%) = \frac{(M_i - M_t)}{M_i} \times 100\% \quad (1)$$

where M_i is initial weight (g) of the film and M_t is final weight (g) of the film.

On the other hand, swelling capacity was measured by cutting the film into dimension of 2 × 2 cm and its weight was measured. The film was dried at 110 °C for one hour, again record the weight (M_o). It was then put in a beaker containing 10 mL of deionized water for about 10 minutes. The excess water was removed by putting it on tissue paper and recording the weight (M_i). The swelling capacity was calculated using Equation 2.

$$Swelling\ capacity\ (\%) = \frac{(M_i - M_o)}{M_o} \times 100\% \quad (2)$$

where M_o is weight (g) of the film after drying at 110 °C and M_i is weight (g) of the film after soaked in water.

Water Vapor Permeability (WVP)

The film was put on the mouth of an Eppendorf centrifuge tube (50 mL), with anhydrous silica gel inside the tube, covering the whole area. The tube was then put in a desiccator containing aquadest (100% RH, 25 °C). The weight of the tube was measured every 24 hours for 7 days straight and calculated using Equation 3 (Liu *et al.*, 2019).

$$WVP = \frac{W \times X}{t \times A \times \Delta P} \quad (3)$$

where W is weight increase of the centrifuge tube (g), X is film thickness (m), t is time needed for the tube weight to increase (s), A is permeation area of the sample (m^2), ΔP is saturated vapor pressure at 25 °C.

Mechanical Properties

Two of the mechanical properties that would be determined were tensile strength and elongation at break. These properties were measured using a texture analyzer (Liu *et al.*, 2019). Each measurement was done triplo and the results were calculated using Equations 4 and 5.

$$Tensile\ strength\ (MPa) = \frac{F}{x \times W} \quad (4)$$

$$Elongation\ at\ break = \frac{\Delta L}{Lo} \times 100 \quad (5)$$

where F is force needed to break the film (N), X is the thickness of the film (mm), W is the width of the film (mm), ΔL is the increased length when the film breaks (mm), and L_o is the initial film length (mm).

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Functional Group and Surface Morphology Analysis

Fourier transform infrared (FTIR) spectrum of the film was recorded using FTIR IRPrestige-21 from 4000 to 400 cm^{-1}. The properties of film surface will be observed using Scanning electron microscopy (SEM) with a magnification of 8000×.

Statistical Analysis

All data are presented as mean ± standard deviation, and the statistical significance was determined using t-test. Significance was determined using p-value of <0.05. A one-way analysis of variance (ANOVA) and Tukey's post hoc test were used for comparisons. Kruskal-Wallis test was used to examine data that are not normally distributed.

RESULTS AND DISCUSSION

Thickness, Water Content, Swelling Capacity and Water Vapor Permeability (WVP)

The thickness of the films increases as the amount of cyclodextrin present in the film solution is higher as can be seen in Table 2. It is suggested that cyclodextrin acts as a cage that envelops carrageenan (Szente and Fenyvesi, 2017), as a result, the more cyclodextrin present in the film matrix, it will prevent the loss of carrageenan, resulting the trend in thickness. The process of spreading the film solution onto the container could also explain the difference in thickness. The gel-like solution made it difficult to pour it completely, hence the difference in the amount that went to the container. In terms of water content, the film with the addition of cyclodextrin presented a lower water content in the film. The hydrophobic nature of inner matrix of cyclodextrin (Karali *et al.*, 2025) could be the reason for these results. Even though the outer matrix of cyclodextrin is hydrophilic (Carina *et al.*, 2021), its circular-like shape could only bind a small amount of water compared to the hydrophilic structure of carrageenan.

Swelling capacity ranges from 21.143 to 31.484. Films that contain a higher level of cyclodextrin exhibit a smaller ability to hold water. Similar to the water content, the structure of cyclodextrin would make it hard for water to adsorb and remain only on the surface. Although carrageenan is a hydrogel-forming compound, water could not reach it, as it is assumed to be enveloped by cyclodextrin. The hydrophilic property of the film could be described by its WVP value (Elewi *et al.*, 2025). The WVP was getting slightly higher when the concentration of cyclodextrin was elevated. Although the difference was small, it could indicate that cyclodextrin creates a condition where water flows harder through the film. These results are in agreement when the nature of cyclodextrin is considered. The hydrophobic state of cyclodextrin inner matrix performs as a barrier for water from getting through the film. It is assumed that the structure of κ-Carrageenan/β-Cyclodextrin is more compact and denser compared to film without cyclodextrin. The addition of cyclodextrin has the potential to produce a better bioplastic for food packaging, as reduced WVP is necessary to make good packaging (Wen *et al.*, 2021). The statistical results, however, assume that there is no difference between samples in all parameters, indicating that the amount of cyclodextrin used in this study was not adequate to show its actual effects. The level of differences of cyclodextrin used should be more diverse.

Table 2. Physical properties of the film.

Sample Code	Thickness (mm)	Water content (%)	Swelling capacity (%)	WVP (10^{-11} g m^{-1} s^{-1} Pa^{-1})
kb-0	0.577 ± 0.0090423[a]	0.242 ± 0.0596237[a]	31.484 ± 1.5154212[a]	11.702 ± 0.0109710[a]
kb-10	0.580 ± 0.0072547[a]	0.177 ± 0.0037009[a]	21.557 ± 9.8355631[a]	11.865 ± 0.0200685[a]
kb-20	0.585 ± 0.0015373[a]	0.155 ± 0.0775227[a]	21.143 ± 6.9173678[a]	11.857 ± 0.0148910[a]

Values are given as mean ± SD (n= 3)

Mechanical Properties

The mechanical properties measured were tensile strength and elongation at break (Table 3). These properties determined the impact of cyclodextrin addition to the film matrix. Tensile strength is described as the maximum stress a material can endure while stretched before it breaks. Tensile strength illustrates the strength of any material, and it is the common property to determine the quality of packaging (Shaikh *et al.*, 2021; Tafa *et al.*, 2023). The film with higher concentration of cyclodextrin has better tensile value. kb-20 had almost doubled of tensile strength compared to kb-0, which did not contain any cyclodextrin. Elewi *et al.* (2025) reported similar results where incorporation of cyclodextrin as a filler improved tensile strength. It is suggested that adding cyclodextrin would

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create stronger polymer-filler interaction and better particle distribution. While other study showed opposite trend, it comes down to microstructural alteration caused by cyclodextrin in the polymer (Elewi *et al.*, 2025).

Another mechanical property is elongation at break, which is defined as the maximum change in length a material can afford before it begins to break. It describes the elasticity of a material, which is one of requirements for a packaging in order for it to be molded into different forms and maintain it during transportation (Shaikh *et al.*, 2021; Tafa *et al.*, 2023). Generally, E% value increases as more cyclodextrin is present in the film matrix. Stronger intermolecular bonds will make the film matrix lose its flexibility. As interaction between carrageenan and cyclodextrin results in more bonds forming, however, the level of cyclodextrin added to the film matrix is not adequate to contribute to such an effect. Initially, at 10%, cyclodextrin is seen to make the bioplastics to be more elastic. However, as the level doubled, its effect is considerably insignificant. Biodegradable film is usually has a low E% compared to conventional thermoplastics (Gabriel *et al.*, 2021; Shaikh *et al.*, 2021). An article published by Elewi *et al.* (2025), stated that cyclodextrin interaction with polymer will create more hydrogen bonds, contributing to stiffness of the film. Increasing level of filler, such as starch produced a film with lower E% as well (Tafa *et al.*, 2023). The level of cyclodextrine need to be more diverse in order to obtain significant conclusion regarding its effect on this particular parameter.

Table 3. Mechanical properties of the film.

Sample code	Tensile Strength (MPa)	Elongation at break (%)
kb-0	5.482 ± 1.3917459[a]	1.647 ± 0.5515323[a]
kb-10	7.981 ± 2.4311261[a]	4.060 ± 1.8612642[a]
kb-20	10.243 ± 1.6563647[a]	4.079 ± 0.7674173[a]

Values are given as mean ± SD (n= 3)

Functional Group Analysis

FTIR spectra of the films are shown in Figure 1. The bands at 925 cm^{-1} represents 3,6-dehydrated galactose repeat unit (stretching of C–O–C bonds), while bands at 846 cm^{-1} describe the galactose-4-sulfate repeat unit (stretching of C–O–S). These bands were present in all graphs and are suspectedly there due to the presence of carrageenan in the film matrix (Barizão *et al.*, 2020). Other bands that showed up are 3400-3500 cm^{-1} (O–H stretching), 2800 – 2929 cm^{-1} (C–H stretching), 1641 – 1649 cm^{-1} polymer bound water, 1159 cm^{-1} (C–C ring), 1259 cm^{-1} (O=S=O asymmetric stretching), and 923 cm^{-1} (C–O–C stretching). These bands are characteristic bonds that can be found in carrageenan and cyclodextrin (Rachmawati *et al.*, 2013; Liu *et al.*, 2019).

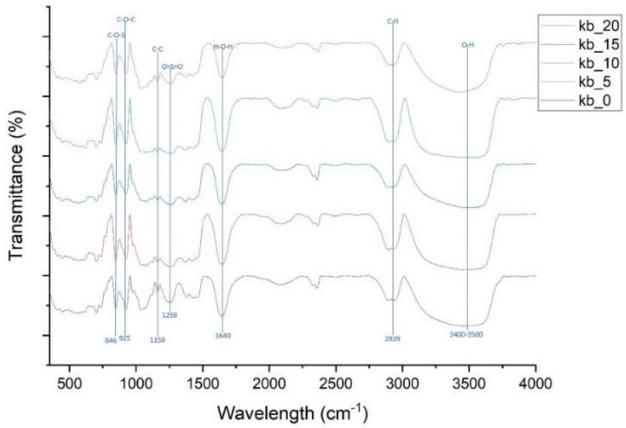


Figure 1. FTIR spectra of κ-Carrageenan/ β-Cyclodextrin (β-CD) film.

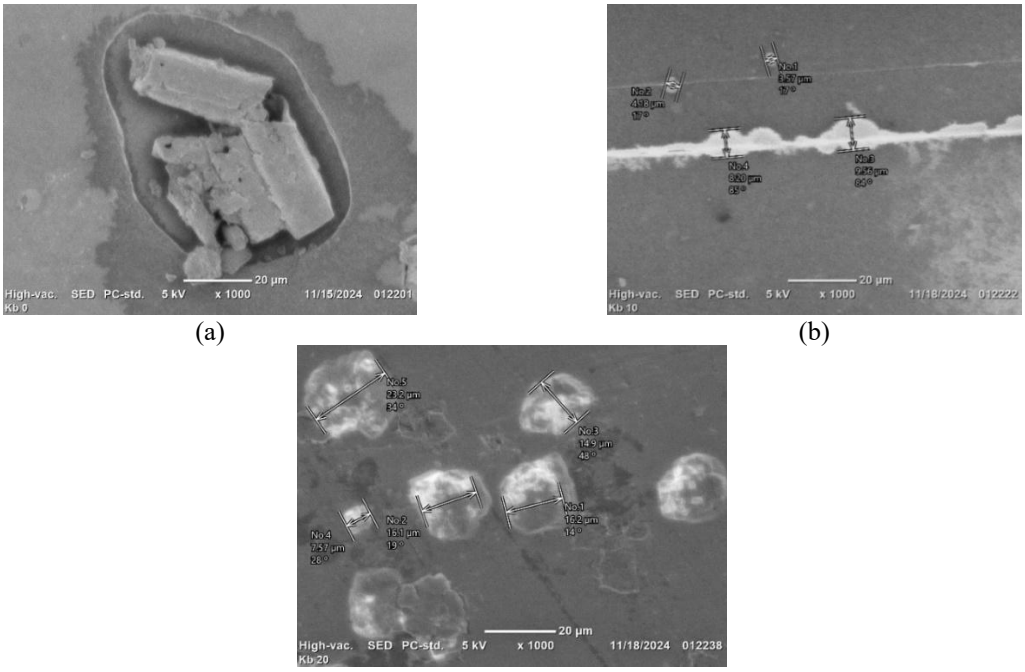
Another characteristic of β-CD was seen at bands of 1640 cm^{-1} (H–O–H deformation bands of water), 1159 cm^{-1} (C–O–C vibration), and 1037 cm^{-1} (C–H, C–O stretching) (Rachmawati *et al.*, 2013). The absorption at 3400 cm^{-1} when β-CD was present was strong and wide, indicating there was a falling bond force due to intermolecular hydrogen bonding (Yuan *et al.*, 2015). The intensity change ranged from 1400 cm^{-1} to 1070 cm^{-1} influenced by hydrogen bond formation (Tian *et al.*, 2024) between κ-Carragernan and β-CD.

Surface Morphology Analysis

The surface of the carrageenan/cyclodextrin was observed using SEM, as can be seen in Figure 2. kb-0, as a control, appeared as rigid, irregular, rough shapes. When cyclodextrin is added, first, a thread-like structure is

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formed (kb-10). It developed a smoother surface as carrageenan was enveloped inside cyclodextrin. At a higher level of cyclodextrin (kb-20), granules, irregular and rough profile is inspected. The round-crystal-like shape which resembles surface without cyclodextrin. These changes are indicating change in its mechanical properties, probably due to interaction between carrageenan and cyclodextrin. For kb-20, the bonds between these two molecules are stronger, leading the structure of the surface to change where some parts joining together.



(a) (b)

(c)

Figure 2. Scanning electron microscope surface of κ-Carrageenan/ β-Cyclodextrin (β-CD) film (a) kb-0, (b) kb-10, and (c) kb-20.

CONCLUSION

Films that constituted from κ-Carragernan and different levels of β-Cyclodextrin were successfully developed. Adding cyclodextrin into the film matrix would cause the thickness of the film, WVP, and tensile strength to rise, while reduced water content, swelling capacity, and elongation at break. At a lower level of cyclodextrin present in the matrix, the surface was smoother and thread-like structure was formed. When the amount of cyclodextrin was higher, it became rougher and form granule-like shape. A suitable level of cyclodextrin needs to be studied in order to obtain a good-quality bioplastic.

CONFLICT OF INTEREST

There is no conflict of interest in this article.

AUTHOR CONTRIBUTION

NN: Methodology, Data Analysis; MDA: Manuscript Review and Editing; NRMA: Methodology; NN: Methodology.

ACKNOWLEDGMENT

This article was made possible by funding from Akademi Komunitas Manufaktur Bantaeng, Ministry of Industry of Republic of Indonesia.

DECLARATION OF GENERATIVE AI

No declaration of generative AI use was provided.

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