JCNaR
Journal of Chemical Natural Resources
Journal homepage: https://jcnar.usu.ac.id

Characterization of Bioplastics from Breadfruit (*Artocarpus altilis*) Starch and Carboxy Methyl Cellulose (CMC) with Glycerol Plasticizer

Cut Fatimah Zuhra*, Suhadiman Dwi Baruna
Department of Chemistry, Faculty of Mathematics and Natural Sciences, Universitas Sumatera Utara, Medan, 20155, Indonesia
*Corresponding Author: cutfatimah@usu.ac.id

ARTICLE INFO

Article history:
Received 8 November 2023
Revised 12 January 2024
Accepted 13 January 2024
Available online 15 January 2024

E-ISSN: 2656-1492

How to cite:

Cut Fatimah Zuhra, Suhadiman Dwi Baruna. Characterization of Bioplastics from Breadfruit (*Artocarpus altilis*) Starch and Carboxy Methyl Cellulose (CMC) with Glycerol Plasticizer. Journal of Chemical Natural Resources. 2024. 5(2):152-158.

https://doi.org/10.32734/jcnar.v5i2.14316

ABSTRACT

Research on the synthesis of bioplastic from breadfruit (*Artocarpus altilis*) starch and carboxy methyl cellulose (CMC) has been successfully carried out with the addition of glycerol. Samples are conducted by varying the glycerol concentrations (10%, 20%, 30%, and 40%) at 60ºC, respectively. The bioplastic product was tested using tensile strength, elongation, water content absorption, biodegradability, and Fourier transform-infra infrared (FT-IR). The tensile strength properties from bioplastic result evaluated is 34.26 N/mm^2. The elongation at break of bioplastic is found at 21.4%. Water content results from bioplastic of 44%. Biodegradability study that bioplastics of 20% in 32 days. Correspondingly, the FT-IR confirmed the functional groups of bioplastics from breadfruit starch and CMC with glycerol.

Keywords: Breadfruit Starch; Bioplastics; Carboxy Methyl Cellulose; Glycerol

ABSTRAK

Penelitian sintesis bioplastik dari pati sukun (*Artocarpus altilis*) dan karboksi metil selulosa (CMC) telah berhasil dilakukan dengan penambahan gliserol. Pengambilan sampel dilakukan dengan memvariasikan masing-masing konsentrasi gliserol (10%, 20%, 30%, dan 40%) pada suhu 60ºC. Produk bioplastik diuji menggunakan kekuatan tarik, elongasi, serapan kadar air, biodegradabilitas, dan fourier transform-infra red (FT-IR). Sifat kekuatan tarik bioplastik hasil evaluasi adalah 34,26 N/mm^2. *Elongation at break* bioplastik ditemukan 21,4%. Kadar air hasil bioplastik sebesar 44%. Studi biodegradabilitas bahwa bioplastik sebesar 20% dalam 32 hari. Sejalan dengan itu, FT-IR mengkonfirmasi gugus fungsi bioplastic dari pati sukun dan CMC dengan gliserol.

Keyword: Bioplastik; Gliserol; Karboksimetil Selulosa; Pati Sukun

1. Introduction

Plastic is a type of packaging often used in everyday life as primary, secondary, and tertiary packaging. The plastic packaging commonly used in daily activities is typically made of synthetic materials. In addition, synthetic plastic cannot be degraded by microorganisms, which are sometimes referred to as non-biodegradable [1]. Consequently, the remaining plastic is usually burned. However, incinerating synthetic plastic trash generates noxious and detrimental gasses, including hydrogen cyanide (HCN) and carbon monoxide (CO), due to incomplete combustion, hence contributing to global warming [2].

Based on data from the Ministry of Environment and Forestry in 2021, the average waste produced by the Indonesian population is around 68.5 million tons. As much as 17 % or 11.6 million tons were occupied by plastic waste. Due to its indispensability in daily life, plastic is necessary. This is demonstrated by the escalating annual utilization of plastic because plastic materials are more practical, lightweight, durable, and cheap compared to wood, paper, and metal materials. Correspondingly, it needs a solution to decrease the utilization of synthetic plastics. Starch is one of the materials that can be used as solution to decrease the proper use of synthetic plastics that can be converted into bioplastic [3].

Starch is a widely available and versatile carbohydrate found in abundance on Earth. Through the utilization of current technology, starch can be transformed into alternative goods, such as bioplastics [4]. One of the starch sources is Breadfruit (*Artocarpus altilis*). Breadfruit is a plant that has a reasonably high starch content, namely 89%. Breadfruit production per hectare averages 4–20 tons in one fruiting season [5]. Based on BPS data (2020), breadfruit production in Indonesia increased in 2020, reaching 190,551 tons compared to breadfruit production in 2019, which was only as much as 122,482 tons. Therefore, it is potentially transformed to be starch and also an essential constituents for the production of bioplastics [3]. Despite its potential as a bioplastic material, there are several disadvantages to consider, i.e., low mechanical and thermal properties and water effects [6]. In order to improve the weakness of starch, some materials need to be added to overcome its weakness, such as carboxymethyl cellulose and glycerol.

Carboxymethyl cellulose (CMC) and glycerol are two materials that can be added to making bioplastics, which also play a role in processing starch into plastic. CMC is a cellulose derivative that is produced by chemically transforming cellulose through esterification and substituting cellulose hydroxyl groups with carboxyl groups. This technique generates molecules with hydrophilic characteristics (capable of attracting water) and a negative charge, rendering them soluble in water [4-5]. Moreover, glycerol is a plasticizer widely used due to its high effectiveness in reducing internal hydrogen bonding, thereby leading to an increase in intermolecular distance. Thus, it is appropriate for the reshaping of hydrophobic plastic materials, primarily starch. Combining these two ingredients is capable of tackling the disadvantages of starch as a bioplastic.

Putra and Saputra (2020), the study examines the impact of including Banana peel (*Musa paradisiaca*) starch and sorbitol on the physical, mechanical, and degrading properties of biodegradable plastic made from muli banana peel starch. Treatment A, which involves adding 1 g of starch and using sorbitol as a plasticizer, yields the most desirable characteristics in biodegradable plastic made from muli-banana peel waste starch based on its mechanical properties. The synthetic banana starch biodegradable plastic possesses the following properties: a thickness of 0.287 mm, a density of 0.667 m/g^3, a water absorption rate of 13.48%, a tensile strength of 13.28 MPa, an elongation of 24.55, and a degradation rate of 18.71% [9]. Hidayat et al. (2013), in their study on biodegradable plastics made from Gembili (*Dioscoreaceae esculenta*) starch, it was found that the best tensile strength was achieved with a 20% of glycerol. The biodegradable plastic exhibited a tensile strength of 12.37 MPa when the starch to CMC ratio was 7:3. The produced biodegradable plastic was analyzed using FT-IR analysis, which revealed the presence of C=O carbonyl groups and C-O ester bonds. It indicates that the plastic is easily degradable in the presence of water and has a high level of biodegradability. [10].

Elean et al. (2018) investigated the production of bioplastics using chempedak seed (*Artocarpus integer*) starch and CMC, with the addition of 10% glycerol. The resulting biodegradable film exhibited a tensile strength of 19.62 MPa, an elongation of 4.98%, a water-absorbed content of 54.33%, and a degradation rate of 39.96% within two days, with a degradability of 24.9 mg/day. FT-IR analysis revealed the presence of O-H alcohol and C-O ether groups, indicating the biodegradable nature of the film. Chempedak seeds contain approximately 36.7% starch, which is relatively low. Therefore, breadfruit can be considered as an alternative to replace chempedak seeds, as it is a commonly used source of starch in Indonesia. [8]. Next, The breadfruit starch has a significant starch concentration, accounting for 76.39% of its entire composition. The remaining 23.61% consists of various substances, including protein, fat, minerals, and water. Simultaneously, breadfruit exhibits amylose and amylopectin levels of approximately 27.76% and 73.24%, respectively. Consequently, the author's research suggests that breadfruit can serve as a viable substitute material for producing bioplastics, thereby addressing the issue of conventional plastic usage in Indonesia [11].

Based on the above, researchers are interested in making bioplastics made from breadfruit starch using variations in glycerol addition. Researchers hope to make environmentally friendly bioplastics that meet the Indonesian National Standard (SNI) specifications.

2. Materials and Methods

The process incorporates the following phases: preparation, synthesis, and characterization. Firstly, breadfruit was obtained from local markets in Medan, Indonesia. Carboxymethyl cellulose (CMC) and glycerol were purchased from Merck (Darmstadt, Germany). Next, the breadfruit was peeled and diced into small parts. Subsequently, the it was pulverized by the blender and then strained to create a pulp. The obtained filtrate was silenced until it formed a precipitate. It was dried under sunlight for 48 hours. The dried precipitate, called starch, was then pulverized, sieved, weighed, and analyzed with an FT-IR spectrophotometer. Secondly, CMC powder and breadfruit starch were dissolved in distilled water and heated on magnetic stirring at 60ºC for 10 minutes. Thirdly, 5% (w/v) of starch solution and a (v/v) glycerol 10% were then homogenized in 500 mL of Beaker glass at 60ºC for 10 minutes and 300 rpm. After that, CMC solution 5% (w/v) was added and homogenized again for 15 minutes until the solution thickened slightly.

Furthermore, the mix solution was poured into an acrylic mold with dimensions of 20 cm x 20 cm. It was dried subsequently at 50ºC for 24 hours. An identical experiment was performed using different concentrations of glycerol (20%, 30%, and 40%). Additionally, FT-IR analysis was conducted to examine the molecular structure. Mechanical properties such as tensile strength and elongation at break were measured to assess the material's capacity to withstand stretching forces. Water absorption and biodegradability tests were also performed to evaluate the material's ibility to absorb water and break down naturally.

3. Results and Discussion

3.1. Tensile Strength Test

Tensile strength test of bioplastic film from breadfruit starch and CMC with glycerol can be clearly seen in Figure 1. Based on that, bioplastic film was tested with UTM RTF 1350, which bioplastic film size of 2 cm x 6 cm clamped at the bottom.

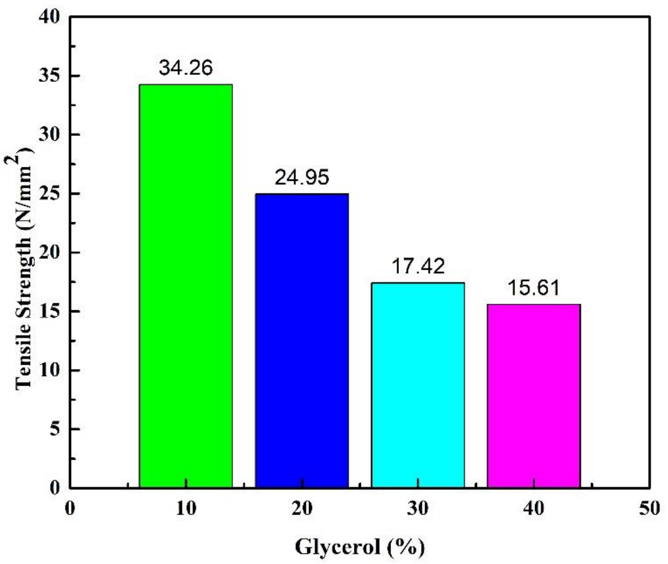

Figure 1. Tensile strength histogram of the bioplastics film of breadfruit starch and CMC with varying of glycerol concentrations

Based on the histogram, the bioplastic from breadfruit starch and CMC with the inclusion of glycerol (10%) encounters the highest value of its tensile strength of 34.26 MPa. In contrast, bioplastic film has a minimum tensile strength value is bioplastic film with adding of glycerol (40%) of 15.61 Mpa. It is due to the glycerol properties that can reduce intermolecular interactions of the starch and CMS. This corresponds to the research by [12], which stated that the increase glycerol concentration as a plasticizer causes decreasing the tensile strength value. Despite on that, Indonesian National Standard (SNI No. 7188:7:2016) declared that the standard of the bioplastic film ranges of 24.7 MPa to 302 MPa. It is indicated that the bioplastic film from breadfruit starch and CMC with adding of glycerol concentration has achieved the required criteria. In other research, which was reported by Warzukni (2020), claimed that bioplastic film based porang (*Amorphophallus muelleri*) starch has a tensile strength value of 20.890 MPa. It noticed that based breadfruit starch and CMC with adding of glycerol can be used as bioplastic film [13].

3.2. Elongation at Break Test

In order to obtain the elongation at break of bioplastic film from breadfruit starch and CMC with glycerol was done with tensile strength procedure, and histogram was displayed in Figure 2. The elongation at break value in the tensile strength test increases as the concentration of glycerol is added, demonstrating an inverse proportionality. As Figure 2 shows the optimum value of elongation at break of bioplatic film from breadfruit starch and CMC with adding glycerol (40%) of 32.6 MPa. At the same time, the bioplastic film (glycerol 10%) has a minimum value of 21.4 MPa. It corresponds the suitability of adding glycerol and making the bioplastic film more elastic. It causes the hydrogen bonds of starch and CMC to be reduced [14]. Therefore, the use of glycerol as a plasticizer must be adjusted due to its impact on the mechanical characteristics of the resulting bioplastic. Corresponding to SNI No. 7188:7:2016, the specification for elongation at break of bioplastic film is specified 21% to 220% [13].

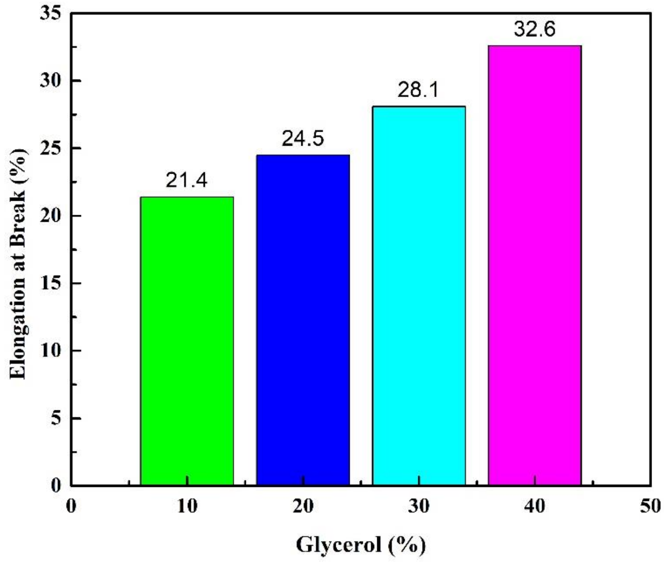

Figure 2. The histogram of the elongation at break of a bioplastics film made from breadfruit starch and CMC, with different concentrations of glycerol.

3.3. Water Content Test

The purpose of the water content test was to analyze the presence and regularity of bonds in bioplastic film made from breadfruit starch and CMC, with the addition of glycerol. This is achieved by measuring the percentage of molecular weight increase resulting from the addition of solvents into the polymers, which leads to the formation of an expanding gel [15]. The degree of swelling was measured by subtracting the mass of the dehydrated bioplastic from the weight of the bioplastic film after soaking with distilled water, dividing it by the grams of the bioplastic film before soaking, and then multiplying the result by 100%. The study conducted water absorption testing to determine the optimal variation. The water absorption test findings are visually represented in Figure 3.

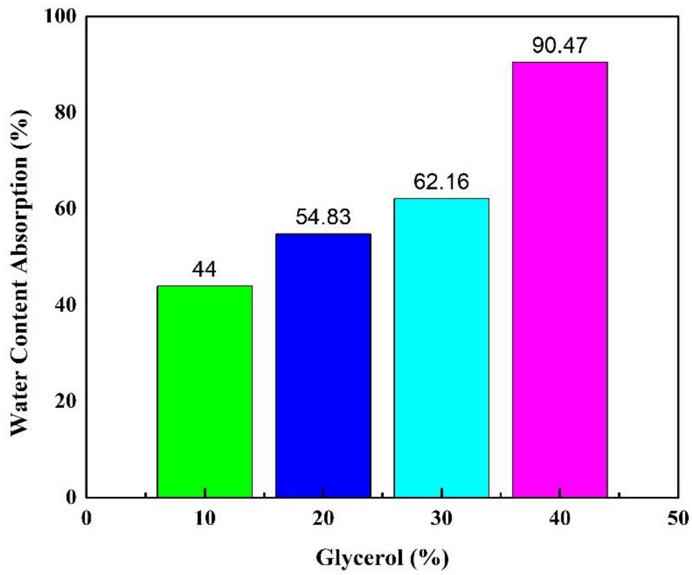

Figure 3. Water content test histogram of the bioplastics film of breadfruit starch and CMC with varying of glycerol concentrations

Figure 3 illustrates that the water content of bioplastic films (glycerol 10%) has absorbed the water of 44%. Bioplastic film (glycerol 40%) has 90.47%. Increasing glycerol concentration which was added into bioplastic film also increase the capacity water absorbed due to the free volume and increase the gap to be occupied water molecules [16].

3.4. Biodegradability Test Results

The synthesized samples' biodegradability are presented in Figure 4. The biodegradability of synthesized samples confirmed a daily decrease in mass. The largest weight loss (%) was found in the bioplastic film with a variation of 10% glycerol. The mass loss was 60.9% on the thirty-second day. Furthermore, the bioplastic film (glycerol 40%) had a mass loss of 77.5%. The degradation of bioplastics can be attributed to the impact of microorganisms present in the compost soil where they are utilized. During the process of biodegradation, complex compounds undergo a thorough transformation into simpler substances, such as water (H=O) and carbon dioxide (CO_2), which are then expelled. [14-15].

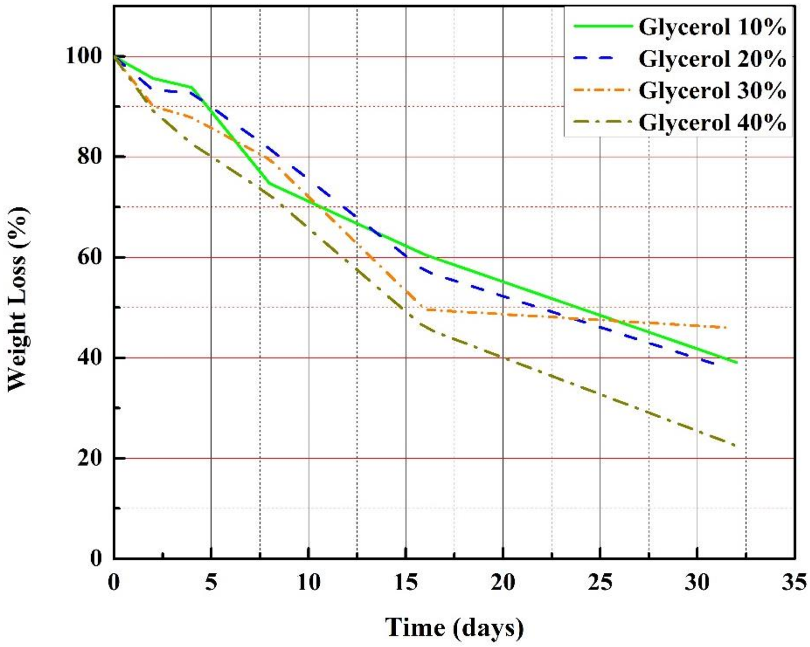

Figure 4. Biodegradability graph of the bioplastics film of breadfruit starch and CMC with varying of glycerol concentrations

The degradation ability of samples will increase due to increasing glycerol. Upon absorption of water, the hydroxyl group present in starch catalyzes the hydrolysis reaction, resulting in the decomposition of the bioplastic into smaller fragments that rapidly degrade in the soil [19]. Chemically, the bioplastic film is biodegradable because it is made from organic and natural raw materials that easily react with water and bacteria, and are sensitive to physical and chemical environmental conditions. The degradation process is inherently affected by factors such as the positioning of the sample, exposure to sunshine, duration of burial, level of humidity, and the presence of microbes. This irregular degradation indicates that soil microorganisms also affect the degradation of bioplastic samples produced [20].

3.5. FT-IR Analysis Results

Analysis of bioplastic films from breadfruit starch and CMC with glycerol used FT-IR were Shimadzu instrument. The objective was to identify any functional group bonds present in the material and determine their relationship to the wave number. As shown in Figure 5, FT-IR spectra of breadfruit starch demonstrate an absorption peak at 3265 cm^{-1}, which confirms the existence of a hydroxyl group (O-H) originating from the α-glucose unit. The C-H stretching band is observed by the peak at 2922 cm^{-1} [21]. On the other hand, the band at 1640 cm^{-1} exhibited C-O cyclic rings from glucose monomer. In addition, wavenumber 1341.8 cm^{-1} investigates C-H bending, as well as C-O-C at 1148 cm^{-1}. Correspondingly, FT-IR spectra of breadfruit starch and CMC with adding glycerol (10%) shows insignificant differences in the peak. The absorption peak is recognized in 1640 cm^{-1} due to C=O, represent of ester. The wavenumber 1587.8 cm^{-1} shows CO bond of the aldehyde, and wavenumber 1341.8 cm^{-1} demonstrated the symmetrical CH of CH_3. Hence, the bioplastic film synthesized from breadfruit starch and CMC with glycerol exhibits biodegradability due to the existence either C=O as well as C-O-C functional groups groups [22].

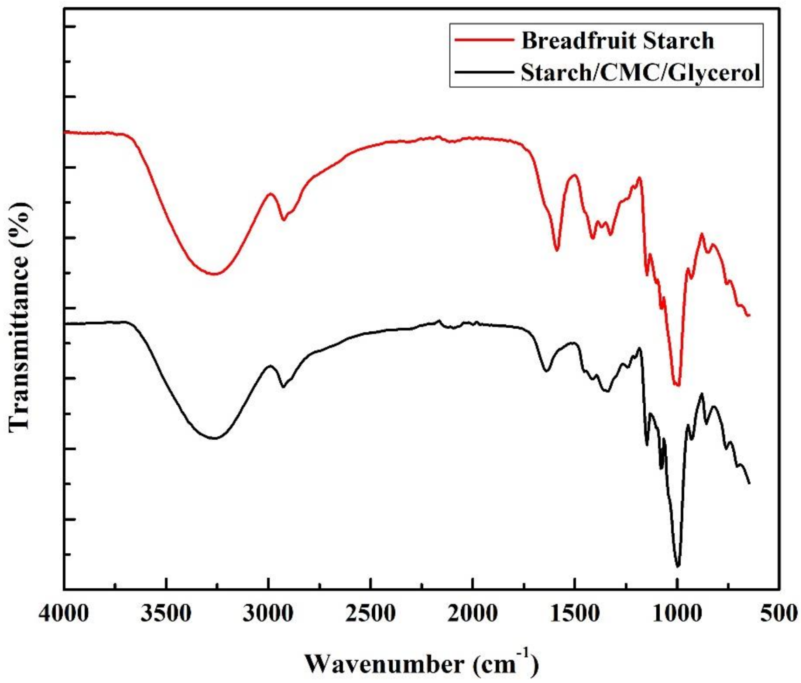

Figure 5. The FT-IR spectra of breadfruit starch and the bioplastics derived from a mixture of breadfruit starch, CMC, and glycerol

4. Conclusion

Synthesis of bioplastic from breadfruit (*Artocarpus altilis*) starch and carboxy methyl cellulose (CMC) has been successfully carried out with addition of glycerol. The addition of glycerol has been shown to impact the properties of the bioplastic film. Based on the findings of the study, it is evident that adding CMC increases the mechanical properties of bioplastic film. In contrast, adding of glycerol increases elastic and water content properties but decreases the mechanical properties. Further study on biodegradability properties showed that bioplastic film produced can be degraded. In other aspects, FT-IR results also indicated functional groups from breadfruit and bioplastic results. Consequently, the bioplastic obtained corresponds to SNI 71887.7:2016 and can can serve as a substitute for traditional plastic.

5. Acknowledgments

The authors acknowledge and appreciate the support and resources offered through the Organic Laboratory of Universitas Sumatera Utara, for example their workspace, scientific competence, and technical assistance.

6. Conflict of Interest

The authors assert to possess no conflicts of interest.

References

[1] M. Borghei, A. Karbassi, S. Khoramnejadian, A. Oromiehie, and A. Hossein Javid, “Microbial biodegradable potato starch based low density polyethylene,” *African J. Biotechnol.*, vol. 9, no. 26, pp. 4075–4080, 2010.

[2] D. Ahmann and J. R. Dorgan, “Bioengineering for pollution prevention through development of biobased energy and materials state of the science report,” *Ind. Biotechnol.*, vol. 3, no. 3, pp. 218–259, Oct. 2007, doi: 10.1089/ind.2007.3.218.

[3] Y. Darni and H. Utami, “Studi Pembuatan Dan Karakteristik Sifat Mekanik dan Hidrofobisitas Bioplastik Dari Pati Sorgum,” *J. Rekayasa Kim. dan Lingkung.*, vol. 7, no. 2, pp. 88–93, 2010.

[4] Z. N. Diyana *et al.*, “Physical properties of thermoplastic starch derived from natural resources and its blends: A review,” *Polymers (Basel).*, vol. 13, no. 9, pp. 5–20, 2021, doi: 10.3390/polym13091396.

[5] I. Farida, K. Syamsu, and M. Rahayuningsih, “Direct bioethanol production from breadfruit starch (*Artocarpus communis forst*) by engineered simultaneous saccharification and fermentation (ESSF) using microbes consortium,” *Int. J. Renew. Energy Dev.*, vol. 4, no. 1, pp. 25–31, 2015, doi: 10.14710/ijred.4.1.25-31.

[6] M. M. Abe *et al.*, “Advantages and disadvantages of bioplastics production from starch and

lignocellulosic components," *Polymers (Basel).*, vol. 13, no. 15, 2021, doi: 10.3390/polym13152484.

[7] N. Kamal, "Pengaruh Bahan Aditif CMC (Carboxyl Methyl Cellulose) Terhadap Beberapa Parameter Pada Larutan Sukrosa," *J. Teknol.*, vol. 1, no. 7, pp. 78–84, 2014.

[8] S. Elean, C. Saleh, and N. Hindryawati, "Pembuatan Film Biodegradable dari Pati Biji Cempedak dan Carboxy Methyl Cellulose dengan Penambahan Gliserol," *J. At.*, vol. 3, no. 2, pp. 122–126, 2018.

[9] E. P. D. Putra and H. Saputra, "Karakterisasi Plastik Biodegradable Dari Pati Limbah Kulit Pisang Muli Dengan Plasticizer Gliserol," *J. Teknol. Pertan. Andalas*, vol. 24, no. 1, 2020.

[10] M. K. Hidayat, Latifah, and S. M. R. Sedyawati, "Penggunaan Carboxy Methyl Cellulose Dan Gliserol Pada Pembuatan Plastik Biodegradable Pati Gembili," *Indones. J. Chem. Sci.*, vol. 2, no. 3, pp. 253–258, 2013.

[11] I. G. Sanjaya and T. Puspita, "Pengaruh Penambahan Kitosan dan Plasticizer Gliserol Pada Karakteristik Plastik Biodegradable Dari Pati Limbah Kulit Singkong," *J. Tek. Kim. FTI-ITS*, 2009.

[12] T. Bourtoom, "Review article edible films and coatings: characteristics and properties," *J. Int. Food Res.*, vol. 15, no. 3, pp. 237–248, 2008.

[13] I. A. Warzukni, "Pembuatan Dan Karakterisasi Plastik Biodegradable Pati Porang Dan Kitosan Dengan Plasticizer Gliserol. Skripsi.," Universitas Sumatera Utara, 2020.

[14] K. Selpiana., Riansya, J. F dan Yordan, "Pembuatan Plastik Biodegradable Dari Tepung Nasi Aking," Palembang, 2015.

[15] M. Kristiani, "Pengaruh Penambahan Kitosan dan Plasticizer Sorbitol Terhadap Sifat Fisiko-Kimia Bioplastik dari Pati Biji Durian (*Durio zibethinus*),"Universitas Sumatera Utara, 2015.

[16] F. Anggarini, "Aplikasi Plasticizer Gliserol Pada Pembuatan Plastik Biodegradable dari Biji Nangka. Skripsi.," Universitas Negeri Semarang, 2013.

[17] M. Fingerman, *Bioremediation of Aquatic and Terrestrial Ecosystems*. New Orleans: Science Publishers, Inc., NH, USA, 2005.

[18] T. Sumarsono, "Biodegradasi Campuran Benzen, Toluen, dan Xilen (Btx) dalam Adsorben Clay oleh Konsorsium Mikroba dengan Penambahan Biosurfaktan Pseudomonas Putida T1(8)," Universitas Airlangga, 2011.

[19] R. Ardiansyah, "Pemanfaatan Pati Umbi Garut untuk Pembuatan Plastik Biodegradable," Universitas Indonesia, 2011.

[20] L. Nurdini, D. Holipah, and R. M. Maulidina, "The Influence of Glycerol as Plasticizer in Physical Properties of Bioplastic from Bread Fruit Starch," in *Seminar Nasional Teknik Kimia "Kejuangan" : K07-1 - K07-6*, Yogyakarta, 2017.

[21] C. F. Zuhra, M. Ginting, and D. S. Siregar, "Synthesis of Cross-linked Breadfruit Starch (Artocarpus communis) using Epichlorohydrin," no. Icocsti 2019, pp. 218–222, 2020, doi: 10.5220/0008878902180222.

[22] K. Wilpiszewska, A. K. Antosik, B. Schmidt, J. Janik, and J. Rokicka, "Hydrophilic films based on carboxymethylated derivatives of starch and cellulose," *Polymers (Basel).*, vol. 12, no. 11, pp. 1–16, 2020, doi: 10.3390/polym12112447.