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Effect Carrageenan to Biodegradable Plastic From Tubers

Toto Rusianto[1,✉], Murni Yuniwati[2], Hary Wibowo[1]

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[1] Mechanical Engineering Department, Institut Sains & Teknologi AKPRIND Yogyakarta, Indonesia 55222
[2] Chemical Engineering Department, Institut Sains & Teknologi AKPRIND Yogyakarta, Indonesia 55222

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Abstract

Plastic waste can cause serious environmental problems. This can be overcome by various efforts; one of them is by replacing the use of conventional plastic with biodegradable plastic. Biodegradable plastic can be produced from tubers containing starch. The purpose of this study was to explain the suitability of two tuber species with typical protein quality and different starch structures. Starch was obtained from suweg tuber (Amorphophallus campanulatus) and ganyong (Canna edulis Ker). The material invested was obtained by dissolving 4 grams of each starch, each of which was dissolved with distilled water then added with various weight carrageenan. The tensile strength of biodegradable plastic was tested using tensile testing machines, biodegradation of soaked plastics using EM4 (Effective Microorganism) with reduced weight measurements, and Fourier Transform Infra-Red (FTIR) was used to identify the structure of compounds contained in biodegradable plastics. The tensile strength test results of biodegradable plastic from ganyong/canna tubers were 3.35 MPa with elongation of 13.51%, while the plastic from suweg tubers of 2.45 MPa with elongation was 13.68% on the addition of 5% carrageenan, respectively. Plastic degradation testing obtained plastic decomposition up to 100% in 37 days for ganyong and 34 days for suweg, respectively. It showed that the plastics was easily degraded. Identified by FTIR showed chemical structures of OH phenolic alcohols, C = O carbonyls, CO esters, NH amides and amines, and C≡C alkyne.

INTRODUCTION

In general, plastic is used as packaging. This is because the shape are elastic, lightweight but strong, not easily broken, transparent and waterproof, but in fact plastic has a negative impact (Ezeoha & Ezenwanne, 2013). Plastic waste can pollute the environment because it takes decades to decompose and can produce dioxin when burned. Based on this problem, it is necessary to use environmentally friendly plastic materials that are easily biodegradable. This material is obtained from biodegradable in the environment, available in large quantities in nature, and can produce products that have the same strength as synthetic plastics (Bourtoom, 2008). Development of biodegradable plastics is one solution to overcome this problem that can replace plastic polymer (Wu & Liao, 2007). Many researchers had conducted research on biodegradable plastics using starch. The starch was obtained from wheat, corn, cassava, which was widely available in agriculture. The starch is the raw material of natural, renewable, cheap, and can naturally break down completely (Wu, 2005). In this report, the biodegradable plastic used starch that was obtained from suweg tubers (*Amorphophallus campanulatus*) and ganyong (*Canna edulis Ker*). Plants of the suweg and ganyong tubers can be seen in Figure 1.

Suweg plants and ganyong are Indonesian local commodity tubers (Richana & Sunarti, 2004;

✉Corresponding author:
Mechanical Engineering Department, Institut Sains & Teknologi AKPRIND Yogyakarta, Indonesia 55222
E-mail: toto@akprind.ac.id

Kasnoet al., 2006). Suweg and ganyong are still not widely known and used by Indonesian people. The use of suweg and ganyong tubers are still limited to research on biodegradable plastic. These tubers have fairly high starch content (Hargono, Santoso, & Deborah, 2016). Therefore suweg tubers and ganyong can be used as the main ingredients in making biodegradable plastics because the carbohydrates contained in the tubers can be taken to produce starch.

(a)

(b)

Figure 1. (a) suweg plants and (b) ganyong/canna.

Biodegradable plastic from starch still has disadvantages so it needs additives to improve its properties. Additives as plasticizers can increase the elasticity of bioplastic materials. Carrageenan and glycerin are additional materials used for the manufacture of biodegradable plastics. Carrageenan is a polysaccharide family of sulfate extracted from red algae types of edible seaweed. Carrageenan is widely used in food and other industries for thickening, swelling and stabilizing properties. Their main application is milk and meat products because these ingredients are strongly bound to food proteins. Glycerin functions as a sweetener and solvent that can help preserve food. Glycerin is also used as an additive in low-fat, commercially made foods (for example, bread), and as a thickening agent in beverages. Glycerin is environmentally friendly and abundant, and non-toxic and can inhibit water evaporation in the product.

MATERIALS AND METHODS

Research on the manufacture of biodegradable plastic from starch obtained from suweg tubers and ganyong with the addition of carrageenan and glycerin plasticizers has not been widely carried out. Therefore, research is needed to make biodegradable plastic from tubers with the addition of carrageenan plasticizers and glycerin to improve the mechanical properties of biodegradable plastics. This research was conducted to determine the use of suweg tubers and ganyong as biodegradable plastic. The results of this study are expected to be useful to provide environmentally friendly bioplastics to reduce the use of conventional plastics so as to overcome plastic waste.

This degraded plastic research used an experimental method with various concentrations of carrageenan and glycerin. The specimens of bioplastic were analysis included of starch content, analysis of water content in starch, tensile strength, percent elongation, biodegradation, and Fourier-changing infrared spectroscopy/FT-IR to identify chemical structure. Stand/retort was used for chemical reactions and processing tools to produce biodegradable plastics that was shown in Figure 2.

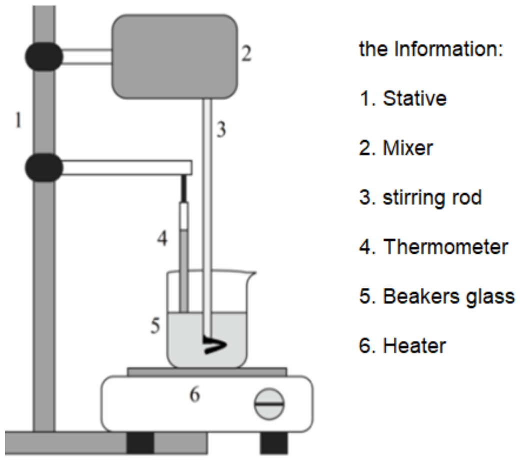


Figure 2. A retort stand for biodegradable plastics processing.

Suweg tubers and *ganyong* were peeled and cut into small pieces and mashed with milling using water to produce colloidal starch. The colloidal starch was filtered to remove water. The precipitate was dried using an oven at 70 °C until the starch completely dry. Then the starch was analyzed. The starch from *suweg* tuber and *anyone* made by dissolving 4 grams into 70 ml of distilled water and adding the mass ratio of carrageenan (0%, 5%, 10%, 15%, and 20% weight) and glycerin 1.3%, 2.0% , 7.0%, 4.0%, 5.3%, and 6.7% volume) in each starch varied, respectively. The mixture was added with acetic acid at a concentration of

5% by volume at 70 °C. Then the mixture was heated at 85 °C and stirred at a speed of 300 rpm for 50 minutes. The process was carried out until liquid thickens. Then, the liquid thickens was poured into a baking dish and dried in an oven with a temperature of 60 °C.

The mechanical properties of biodegradable plastics tested include tensile strength and elongation. The test was carried out with ASTM 882-91 procedur (Husain, Rahman, Senawi, & Khairudin, 2016). The tensile strength and extension were measured using Universal Testing Instrument (UTI). Biodegradation test served to determine the durability of biodegradable plastic can be described. The test was carried out by means of the biodegradable plastic in size 3 × 3 cm^2 and then put into a porcelain cup with bio-activator EM-4. The specimens were observed until the biodegradable plastic changed and was completely damaged or destroyed for several days. Weight loss from specimens decomposed by bio activators showed a decrease in the rate of degradation of biodegradable plastic materials. Bioplastic structure compounds from *Suweg* and *ganyong* were analyzed using FTIR. Chemical structures contained in plastics can be known using FTIR spectra. The spectra of this constituent element were compared with spectra in starch from *suweg* tubers and *ganyong,* so it can be estimated the type of element interactions that occur.

RESULTS AND DISCUSSION

Content and Test of Moisture

The process of making suweg tuber starch and ganyong contains 7.67% and 6% fine powder starch, respectively. Water content testing with gravimetric analysis was obtained with water content in 11.60% suweg tuber starch and 10.38% ganyong tuber. The value of water content in suweg starch and ganyong according to SNI 01-3451-1994 is a maximum of 15%, so the water content of starch suweg and ganyong produced is still in accordance with the standards. Water content influences starch resistance as a base material for biodegradable plastics (Salmoral et al., 2000). The higher the water content in starch, the shorter the starch resistance so that it will be contaminated more quickly by microbes so that it is not recommended being stored for a long time. According to Setiani et al. (2013) the difference in water content in starch caused by different drying processes, both the method and drying time can significantly affect the amount of moisture content of dry starch. Other factors that influence the amount of water content in starch as explained by Setiani et al. (2013), which states that other factors that affect the humidity of the surrounding air are related to the storage of materials, material properties, and the type of material and treatment that has been experienced by the material (Setiani et al., 2013).

Mechanical Properties

The edible film layer is formed from suweg tuber starch and ganyong with variations in the ratio of carrageenan mass and glycerin volume. The effect of carrageenan on the mechanical properties of biodegradable plastic with tensile strength values can be seen in Figure 3.

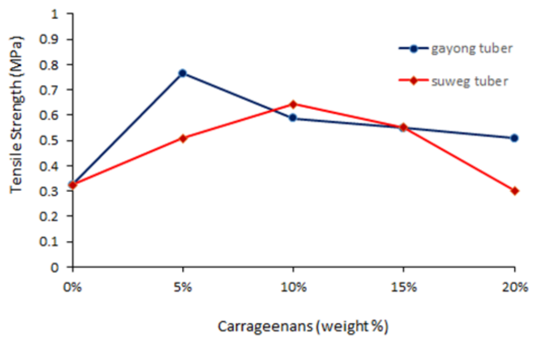


Figure 3. The mass ratio of carrageenan to the value of the tensile strength of the biodegradable plastic.

Increasing the mass ratio of carrageenan to suweg tuber and ganyong with optimum tensile strength of 10% in suweg and 5% in ganyong. This causes the bond between biodegradable plastic constituent molecules to increase, resulting in increasingly complex biodegradable plastic solutions characterized by increasingly high tensile strength. Because carrageenan is able to form a strong polymer matrix and makes the tensile strength between molecules stronger in biodegradable plastics (Handito, 2011). However, in the carrageenan mass ratio of 15% and 20% the value of tensile strength decreases because more and more dissolved solids so that the viscosity of the solution becomes high which causes the stirring process to be less optimal so that the value of tensile strength decreases. The effect of carrageenan on the extension of the specimen can be seen in Figure 4.

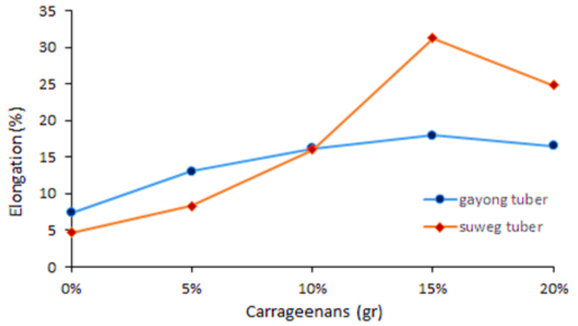


Figure 4. Graph of the relationship of weight ratio of carrageenan and weight of starch to percent elongation.

Figure 4 shows that the greater the use of carrageenan mass, the greater the percentage of biodegradable plastic elongation. That causes the interaction between carrageenan molecules to cause the bioplastic to become more elastic so that the elongation level will increase. According to Krochta and Johnson (1997), the percentage value of the extension is bad if it is less than 10% (Handito, 2011). Biodegradable plastic with a good variation in the mass ratio of carrageenan to starch because it has an elongation percentage value of more than 10%. However, in the variation of carrageenan weight ratio of 0.20, there was a decrease in percent elongation because the addition of excess carrageenan would produce residues from carrageenan which could weaken the building structure of biodegradable plastic so that the percent extension in plastic produced decreased.

Effect of Glycerin to Biodegradable Plastics

Effect of glycerin volume on starch in Tensile Strength Value. To study the effect of adding glycerin volume on the study, it was carried out by using a comparison of the volume of glycerin with various starches.

Figure 5 shows that an increase in the volume of glycerin plasticizer to starch, that the value of tensile strength decreases. The decrease in tensile speed caused by the addition of glycerin plasticizers will reduce the internal hydrogen bond in the bonds between molecules, thereby reducing the stability of the solid dispersion system; as a result, the resulting biodegradable plastic has a weakness. The addition of plasticizers will cause a decrease in the tensile force between the polymers in terms of water evaporation which results in a decrease in resistance to the mechanical treatment of biodegradable plastics. According to Lieberman and Gilbert (1973), plasticizers can change the physical properties of biodegradable plastics by reducing the cohesion and mechanical resistance of polymer chains (Lieberman & Gilbert, 1973). Shaikh et. al. (2019) stated the same report, that plasticizers could cause a reduction in internal hydrogen bonds and weaken intermolecular attractions of adjacent polymer chains, thereby reducing the strength of biodegradable plastics (Shaikh et al., 2019). The highest tensile strength values were found from suweg tubers and ganyong in the glycerin plasticizer volume ratio of 1.3%, respectively 2.46 MPa and 3.34 MPa. The effect of glycerin volume with the percentage of extension can be seen in Figure 6.

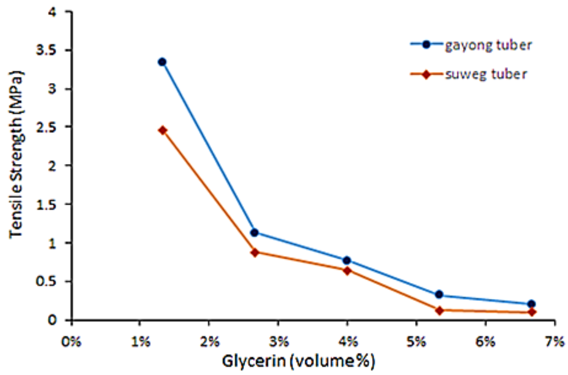


Figure 5. Graph of the relationship between the volume of glycerin and the mass of starch to the value of tensile strength.

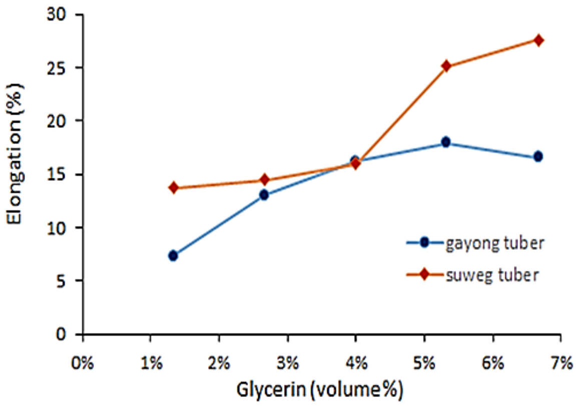
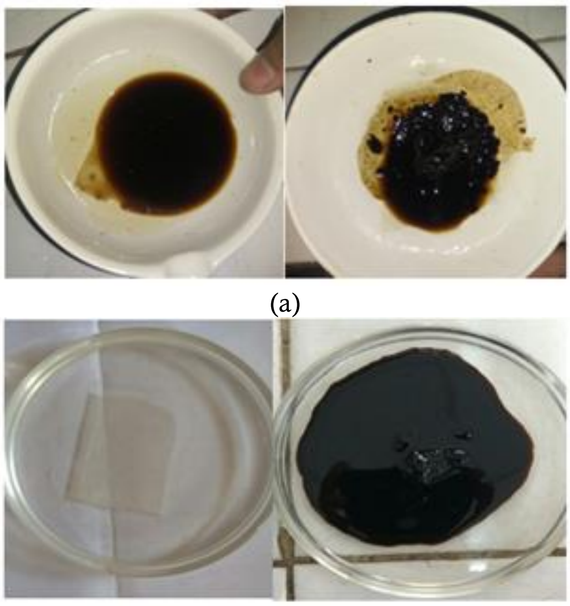


Figure 6. Graph of the relationship of the volume ratio of glycerin to the mass of starch to percent elongation.

From the analysis of percent elongation, the highest yield was obtained by using a glycerin volume ratio of 6.7% from 27.55% in suweg and 16.52% in ganyong. The greater the ratio of the volume of glycerin plasticizers to the mass of additional starch will increase the percent of the yield of biodegradable plastic produced. This is because an increase in the number of plasticizers will reduce the force between molecules, so the level of mobility between molecular chains increases. The result is in an OH group in glycerin, which forms bonds between molecules with a reduced polymer chain. The increase in glycerin will reduce the cohesion bond between the polymers that make up biodegradable. Bourtoom (2008) has reported that glycerin as a plasticizer reduces internal hydrogen bonds by increasing the empty space between molecules, which will be filled by glycerin (Bourtoom, 2008). Likewise, Naderizadeh et.al., (2019) reported that Poly(vinyl alcohol)/PVA plasticizers will increase the flexibility of the biodegradable plastic produced, where more glycerin was added to some extent to make biodegradable plastics formed more elastic and flexible (Naderizadeh et al., 2019).

Biodegradation Test Results

Biodegradation test was carried out to determine the time needed for biodegradable plastic samples to experience degradation. Samples soaked in EM-4 (Effective Microorganism) solution until decomposed. The Experiments can be seen in Figure 7.



(a)

(b)

Figure 7. Biodegarable plastic before degradation (left) and after degradation (right) (a) suweg and (b) ganyong.

The bioplastics that were tested by using EM-4 for degradation showed degraded samples up to 34 days for suweg and 37 days ganyong, respectively. EM-4 contains bacteria that are used for fermentation of the Lactobacillus genus, photosynthetic bacterial actinomycetes, phosphate solvent bacteria, and yeast. This biodegradation process was through anaerobic and aerobic processes. According to Shit & Sha (2014), degradation of polymers is used to express physical changes due to chemical reactions that include breaking the bonds of biodegradable plastic molecules from macromolecules (Shit & Shah, 2014; Hardjono et al., 2016).

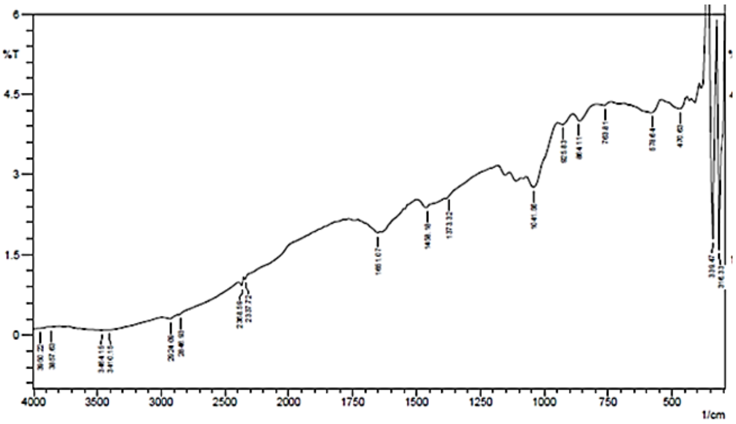
When degradation occurs, hydrolysis processing occurs in a polymer matrix with more hydroxyl groups decomposing faster into smaller pieces. The form of suweg tuber and ganyong starch is a natural polymer that is easily degraded by bacteria. This report is strengthened by Diova's et al. (2013), biodegradable plastics made from starch can be easily degraded by bacteria to break down polymer chains into monomers (Diova et al., 2013). Biodegradable plastic is made from starch with the addition of carrageenan, glycerin as a plasticizer, and acetic acid as a cross agent has proven to be environmentally friendly when compared to synthetic plastics that have only been degraded for 50 years.

FT-IR Identification Result

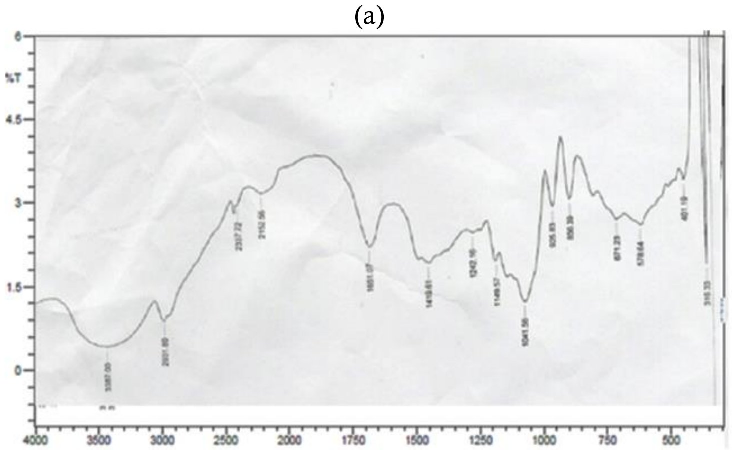
Pieces of bioplastic specimens were identified by FT-IR with differential refractive index, infrared to determine the bond that occurs in the biodegradable plastic identification produced. The results of Absorption bands for the plastic identification by using FTIR were shown in Figure 8.

Table 1 shows the results of the FTIR test which shows several chemical structure ds. Biodegradable plastic has a double bond of chemical structure C = O which is the structure of compounds in carbohydrates shown in wave number 1651.07 cm^{-1}. Carbohydrate molecules are compounds that contain the structure of carbonyl groups (as aldehydes and ketones) and long hydroxyl chains. The O-H chemical structure with the name of the alcohol compound, and the phenol at absorption band of 3464.15 cm^{-1} has a strong effect on the characteristics of the biodegradable plastic produced.

The single Amide (N-H) bond was found in absorption bands of 3410.15 cm^{-1} in this spectra, there is also a group of amine compounds (intermediate uptake at 3500 and Amide configured) (Handayani & Wijayanti, 2015).



(a)



(b)

Figure 8. Spectra produced from FTIR testing results for bioplastics from (a) suweg tuber and (b) ganyong.

Table 1. List of absorption bands spectra of bioplastic identified.

Absorption bands (cm^{-1})		Chemical structure	assignment
suweg	ganyong		
1651.07	1651.07	C=O	carbonyl
3464.15	3387	O-H	Alcohol and phenol to hydrogen
3410.15	3387	N-H	Amide
1041.56	1149.57	C-O	Ester
3410.15	3387	N-H	Aromatic amine
1651.07	1651.07	C=C	Aromit alkene
2368.59	2152.07	C≡ C	Alkyne

Number wave 1041.56 cm^{-1} is a C-O functional group with the name ester compound which shows that the film layer is hydrophilic. The peak with wave numbers 1651.07 cm^{-1} is the functional group C = C, while at the peak of 2368.59 cm^{-1} alkyne compounds are formed with three carbon double bonds (Nigar et al., 2019). FTIR test results show that all groups contained in plastic produced can be degraded. This is in accordance with Darni (2009), that in addition to the functional groups of another hydroxide (OH) functional groups contained in biodegradable plastics are carbonyl (C=O) and ester groups so that with this functional group biodegradable plastics can be degraded. This study found that the O-H, C=O, and ester functional groups (Cifriadi et al., 2011) that produce biodegradable plastic can be degraded (Wu & Liao, 2007).

CONCLUSION

Increasing the mass ratio of carrageenan to starch from suweg tuber and ganyong with the optimum tensile strength of 10% in suweg and 5% in ganyong. Increasing the volume of glycerin plasticizer in starch decreases its tensile strength.

The biodegradation test results showed that biodegradable plastic was degraded or decomposed in 34 days for suweg and 37 days for ganyong. Identify by using The FT-IR (Fourier Transform Infra Red) for the plastic specimens showed that biodegradable plastic was composed by hydrocarbons that are easily biodegradable in the environment.

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REFERENCES

Bourtoom, T. 2008. Plasticizer effect on the properties of biodegradable blend film from rice starch-chitosan. Songklanakarin Journal of Science & Technology. 30(1): 149-165.

Cifriadi, A., Budianto, E., Alfa, A. 2011. Karakteristik Karet Siklo Berbasis Lateks Karet Alam Berbobot Rendah. Indonesian Journal of Natural Rubber Research. 29(1): 35-48.

Diova, D. A., Darmanto, Y., Rianingsih, L. 2013. Karakteristik edible film komposit semirefined karaginan dari rumput laut Eucheuma cottonii dan beeswax. Jurnal Pengolahan dan Bioteknologi Hasil Perikanan. 2(4): 1-10.

Ezeoha, S., Ezenwanne, J. 2013. Production of biodegradable plastic packaging film from cassava starch. IOSR Journal of Engineering. 3(10): 14-20.

Handayani, P. A., Wijayanti, H. 2015. Pembuatan Film Plastik Biodegradable dari Limbah Biji Durian (*durio zibethinus murr.*). Jurnal Bahan Alam Terbarukan. 4(1): 21-26.

Handito, D. 2011. Pengaruh Konsentrasi Karagenan Terhadap Sifat Fisik Dan Mekanik Edible Film. Argoteksos. 21(2-3): 151-157.

Hardjono, H., Permatasari, D. A., Sari, V. A. 2016. Pengaruh Penambahan Asam Sitrat Terhadap Karakteristik Film Plastik Biodegradable Dari Pati Kulit Pisang Kepok (*Musa Acuminata Balbisiana Colla*). Jurnal Bahan Alam Terbarukan. 5(1): 22-28.

Hargono, H., Santoso, A. W., Deborah, G. K. 2016. Pemanfaatan Umbi Suweg (*Amorphophallus sp*) sebagai Bahan Baku Pembuatan Bioetanol melalui Proses Fermentasi dan Distilasi. Paper presented at the Seminar Nasional Teknik Kimia Kejuangan.

Husain, H., Rahman, A. Y., Senawi, N., & Khairudin, S. (2016). Biodegradable Film from Pleurotus Sajor-Caju Waste. Jurnal Teknologi, 78(11-2).

Kasno, A., Trustinah, M. A., Swasono, B. 2006. Prospek Suweg Sebagai Bahan Pangan Saat Paceklik. Prosiding Seminas Nasional Balai Penelitian Tanaman Aneka Kacang dan Umbi 2006. 257-262.

Lieberman, E., Gilbert, S. 1973. Gas permeation of collagen films as affected by cross-linkage, moisture, and plasticizer content. Journal of Polymer Science: Polymer Symposia. 41(1): 33-43.

Naderizadeh, S., Shakeri, A., Mahdavi, H., Nikfarjam, N., Taheri Qazvini, N. 2019. Hybrid Nanocomposite Films of Starch, Poly (vinyl alcohol) (PVA), Starch Nanocrystals (SNCs), and Montmorillonite (Na-MMT): Structure–Properties Relationship. Starch-Stärke. 71(1-2): 1800027.

Nigar, A., Shabbir, M., Akhter, Z., Sabahat, S., Fatmi, M. Q., Bolte, M., Ahmad, I., Janjua, N. K., Mehmood, S. 2019. Synthesis, characterization, docking and electrochemical studies of nitroaromatic amides. Journal of Molecular Structure, 1176(January): 791-797.

Richana, N., Sunarti, T. C. 2004. Karakterisasi sifat fisikokimia tepung umbi dan tepung pati dari umbi ganyong, suweg, ubi kelapa dan gembili. Jurnal pascapanen. 1(1): 29-37.

Salmoral, E., Gonzalez, M., Mariscal, M., Medina, L. 2000. Comparison of chickpea and soy protein isolate and whole flour as biodegradable plastics. Industrial Crops and Products. 11(2-3): 227-236.

Setiani, W., Sudiarti, T., Rahmidar, L. 2013. Preparasi dan karakterisasi edible film dari poliblend pati sukun-kitosan. Jurnal Kimia Valensi. 3(2): 100 – 109.

Shaikh, M., Haider, S., Ali, T. M., Hasnain, A. 2019. Physical, thermal, mechanical and barrier properties of pearl millet starch films as affected by levels of acetylation and hydroxypropylation. International Journal of Biological Macromolecules. 124(March): 209-219.

Shit, S. C., Shah, P. M. 2014. Edible polymers: Challenges and opportunities. Journal of Polymers. 2014: 1-13.

Wu, C.-S., Liao, H.-T. 2007. Study on the preparation and characterization of biodegradable polylactide/multi-walled carbon nanotubes nanocomposites. Polymer. 48(15): 4449-4458.

Wu, C. S. 2005. Improving polylactide/starch biocomposites by grafting polylactide with acrylic acid–characterization and biodegradability assessment. Macromolecular Bioscience. 5(4): 352-361.