Jusami
Homepage: http://jusami.batan.go.id
Jurnal Sains Materi Indonesia
Akreditation No : 36/E/KPT/2019
Date 13 Dec 2019
ISSN 1411-1098
E-ISSN 2614-087X

PROPERTIES OF EDIBLE FILM FROM MODIFIED SAGO STARCH PRECIPITATED BY BUTANOL

Riska Surya Ningrum[1], Dewi Sondari[1], Putri Amanda[1], Bernadeta Ayu Widyaningrum[1], Dian Burhani[1], Fazhar Akbar[1] and Yulianti Sampora[2]

[1]Research Center for Biomaterial, Indonesian Institute of Science - Cibinong Science Center
Cibinong, West Java, 16911
[2]Research Center for Chemistry - Indonesian Institute of Science
Kawasan Puspiptek Serpong 15314, South Tangerang, Banten
E-mail: riska_suryaningrum17@yahoo.com

Received: 27 April 2020 *Revised: 3 July 2020* *Accepted: 10 July 2020*

ABSTRACT

PROPERTIES OF EDIBLE FILM FROM MODIFIED SAGO STARCH PRECIPITATED BY BUTANOL. The edible film is a thin layer that can be used as food packaging and safe for consumption. Starch is a material that can be used as an edible film because it is biodegradable, non-toxic, able to form a strong and transparent film. In this research, sago starch has been precipitated using butanol for 1, 2, 3, 4, and 5 hours. Precipitated sago is reacted with 3% glycerol and then formed into a film by printing it on a petri dish. Characterizations of edible films are FTIR, contact angle, water solubility, swelling power, WVTR, and mechanical properties (thickness, tensile strength, and elongation). Edible film (B2) has the best mechanical properties, that is high hydrophobicity (contact angle is 60,351 degree), tensile strength (6,8843 N/mm^2), and elongation (49,5081 %), also low water solubility (0,3352 %), moisture content (18,6005 %), and WVTR (0,02986 g s^{-1} m^{-2}).

Keywords: Edible film, Sago starch, Precipitation, Butanol

ABSTRAK

SIFAT EDIBLE FILM YANG TERBUAT DARI MODIFIKASI PATI SAGU YANG DIPRESIPITASI DENGAN BUTANOL. Edible film merupakan lapisan tipis yang digunakan sebagai pembungkus makanan dan aman untuk dikonsumsi. Pati merupakan bahan yang dapat dimanfaatkan sebagai edible film karena bersifat *biodegradable*, tidak toksik, mampu membentuk film yang kuat dan transparan. Pada penelitian ini, pati yang digunakan adalah pati sagu yang telah dipresipitasi selama 1, 2, 3, 4, dan 5 jam dengan pelarut butanol. Sagu yang telah dipresipitasi direaksikan dengan gliserol 3% lalu dibentuk menjadi film dengan cara dicetak pada petri dish. Karakterisasi edible film meliputi FTIR, sudut kontak, kelarutan dalam air, swelling power, WVTR, dan sifat mekanik (ketebalan, kekuatan Tarik, dan elongasi). Sampel edible film (B2) memiliki sifat mekanik terbaik, yaitu hidrofobisitas yang tinggi (ditunjukkan oleh nilai sudut kontak sebesar 60,351 derajat), kekuatan tarik dan elongasi yang besar (6,8843N/mm^2 dan 49,5081%), kelarutandalam air, kadar air, dan WVTR yang rendah (0,3352 %, 18,6005 %, dan 0,02986 g s^{-1} m^{-2}).

Kata Kunci: Edible film, Pati sagu, Presipitasi, Butanol

INTRODUCTION

The edible film is a thin layer (film) made of materials that are safe for consumption, such as hydrocolloids (starch, protein, cellulose, alginate, and polysaccharide), lipid (waxes, glycerol, fatty acid), and composites (a mixture of lipid and hydrocolloid components) [1]. The functions of edible film are protecting food from the surrounding environment (water vapor, gas, light, microorganism) and also preventing mass transferred so that the food quality can be maintained.

Starch is one of the materials that can be used as an edible film. Starch is a polysaccharide consisting of amylose and amylopectin. Starch can be obtained from cassava (tapioca starch), sago plants (sago starch), maize, and other tubers whose very abundant resources in Indonesia. Starch can be used as a coating agent that is safe for consumption (edible) because it is biodegradable, non-toxic, able to form a strong and transparent film [2].

Research and development of edible film based on starch had been studied since the last decade. Starch-based edible films have several drawbacks, that is low barrier properties against gases (O_2, CO_2, ethylene) and water vapor due to the presence of pores and crevices causing high permeability [3]. Therefore, it is necessary to make efforts to enhance the mechanical properties, hydrophobicity, and antimicrobial activity of edible films by blending them with other polymers, modifying starch or reducing the particle size of starch.

In the development of edible film starch-based, there is some biopolymer that can be used to blending processes, such as carboxymethyl cellulose nanoparticle [4], polyvinyl alcohol (PVA) [5], chitosan [6], orange peel-oil [7], etc.

Modification process and reducing the particle size of starch can be done by physical, chemical, enzymatic or combination of treatments. Sondari *et al*. [8] had modified sago starch using oxidation, acetylation, crosslink, and ethanol precipitation treatment to improve the contact angle and decrease the moisture content of the edible film.

Among the various technique to modify sago starch, precipitation is a technique that easier, simpler, inexpensive, and need less time to do. Therefore, the aims of this research are preparation and characterization of edible films made from modified sago starch using butanol precipitation technique. Edible film is made by blending modified sago starch with glycerol as a plasticizer. The reason for choosing sago as a material for making edible films is because of its abundant availability in Indonesia. Besides, the development of an edible film based on modified sago starch using butanol precipitation has not been widely used. Characterizations of edible films are functional groups using FTIR, contact angle, water solubility, swelling power, Water Vapor Transmission Rate (WVTR), mechanical properties such as thickness, tensile strength, and elongation.

EXPERIMENTAL METHOD

Materials and Instruments

Sago starch, butanol (Merck), glycerol (Merck), distilled water, Particle Size Analyzer (PSA), FTIR Perkin Elmer, Universal Testing Machine (UTM) Shimadzu AG-IS 50 KN.

Method and Procedure

Modified Sago Starch using Butanol Precipitation Method

4 g sago starch dissolved into 75 ml of distil water, heated at 50 Celsius degree while stirring at 350 rpm for 1 hour, then cooled it in the room temperature. After cooling, the samples reacted with 75 ml dropwise of butanol while stirring at 700 rpm for a certain time (1, 2, 3, 4, and 5 hours). Furthermore, it is centrifuged to separate sago from butanol. The obtained of sago is dried using a freeze dryer. The modified sago was then analyzed for its particle size using Particle Size Analyzer (PSA).

Preparation of Edible Film

4 g sample dissolved into 40 ml distilled water using an ultrasonicator. Add glycerol 3% into it then heated at 105 Celsius degree while stirred at 350 rpm for 30 minutes. Take 10 g and pour it into a petri dish (diameter 9 cm) then dried in the oven at 50 Celsius degree for 24 hours.

Characterization of Edible Film

Characterizations of edible film were functional groups using FTIR at a wavenumber of 4000-400 cm^{-1}, contact angle, water solubility, and swelling power were determined by Yuniar *et al.* [9], Water Vapor Transmission Rate (WVTR) were determined by Othman *et al*. [10] and mechanical properties such as thickness, tensile strength, and elongation of samples were determined using Universal Testing Machine (UTM).

RESULT AND DISCUSSION

Modified Sago Starch using Butanol Precipitation Method

The modification process of sago starch have done by precipitation method using butanol as a solvent with a variation of time: sample B1 for one-hour, B2 for two-hours, B3 for three-hours, B4 for four-hours, and B5 for five-hours precipitation.

The purpose of modification process is to reduce the particle size of sago starch. The small particle size of starch will be enhanced the ability of edible film to block the entry of gas and water vapor so it can maintain the food quality [11]. The particle size and yield of modified sago using butanol can be seen in Table 1.

Table 1 shows that there is no correlation between time of sago precipitation using butanol and the particle size of modified sago. Sample B4 has the smallest particle size, which is 23,2472 μm while B1 has the largest particle size, which is 37,4354 μm.

Table 1. Particle size and yield from modification sago starch process.

Sample Code	B1	B2	B3	B4	B5
Particle size (µm)	37.4354	30.3187	30.3076	23.2472	32.8823
Yield (%)	89.9700	93.2890	86.0253	90.2551	93.8933

Generally, the yield of modified sago that obtained from the butanol precipitation process is relatively large.

Preparation of Edible Film from Modified Sago Starch

Edible films made from native or modified sago with added 3% of glycerol. The function of glycerol is as a plasticizer. Adding glycerol into modified sago solution can make the edible film formed less brittle. According to Togas et al. [12], glycerol is the most widely used as a plasticizer because glycerol is a clear compound, easily dissolves in water, increases the viscosity of the solution, and binds water.

The results of functional group analysis using FTIR can be seen in Figure 1 and Table 2. Generally, the functional groups found in the native sago are similar to modified sago, however, both of them have different absorption intensity of hydroxyl (-OH) groups. All of the edible films, except B5, from modified sago based have lower intensity of hydroxyl group than the edible film from native sago (KO) based.

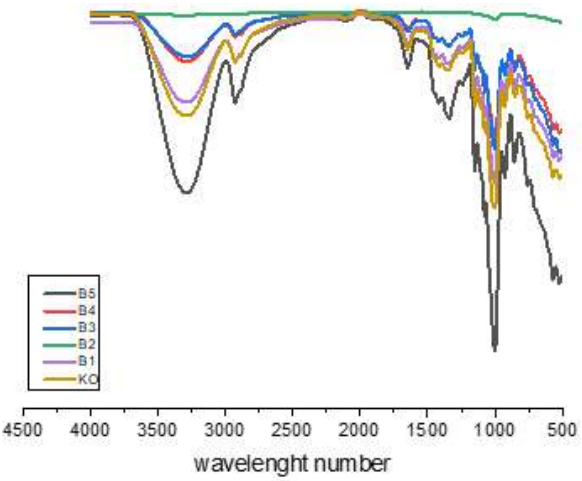


Figure 1. **FTIR spectra of native and modified sago**

Table 2. Functional groups containing in the edible film.

Wavelength Number (cm-1)	Functional Group
3288-3391	OH stretching
2927-2929	C-H stretching
1642-1649	C=O bonding
996-1002	C-O-C complex

Figure 1 shows that sample B5 has the highest hydroxyl group absorption intensity while the B2 has the lowest one. It means that the hydrophobicity of sample B2 is the highest than other samples. In this study, the hydrophobicity characteristic of the edible films was also indicated by the contact angle, moisture content, WVTR, and water solubility result analysis.

The aim of contact angle measurement on a film is carried out to determine the properties of surface material, that is hydrophilic or hydrophobic. The greater the contact angle, the surface material (film) is more hydrophobic.

Similar to FTIR analysis, all edible films, except B5, that obtained from modified sago have higher contact angel than the edible film from native sago (KO) based (Figure 2). It means that edible films obtained from modified sago were more hydrophobic than the edible film from native sago. The result of Sondari *et al*. [8] showed that biofilm obtained from modified sago using crosslinks, acetylation, oxidation, and precipitation method have a higher contact angle and lower moisture content than biofilm obtained from native sago. Modification of sago starch using chemical, mechanical, and enzymatic treatment can be improved the hydrophobic properties of sago [3]. When the hydrophobicity of modified sago starch was increasing, the contact angle of the film obtained from it was also increasing. Figure 2 showed that sample B2 has the highest contact angle, which is 60,351 degrees while B5 has the lowest contact angle, which is 43,887 degrees.

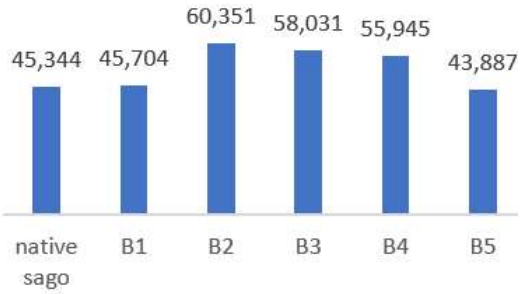


Figure 2. Contact angle analysis of edible film from native and modified sago.

Moisture content analysis is also indicated the hydrophobicity of film and affect the film ability to absorb the water. The more hydrophobicity of film, the lower the moisture content of it. Therefore it can be decreased the quality of the film into a shorter service life due to bacterial interference [13]. In this research, the moisture content of edible film obtained from modified sago starch (except B5) have a lower moisture content than edible film obtained from native sago. B2 has the lowest moisture content than other edible films, which is 18,6005 % (Figure 3).

Water Vapor Transmission Rate (WVTR) analysis was performed to determine the water permeability of the film. The edible film quality will increase with the



Figure 3. Moisture content analysis of edible film from native and modified sago.

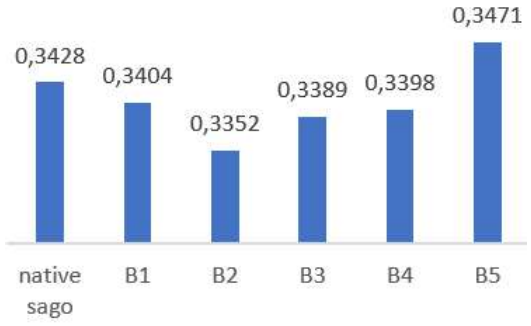


Figure 6. Water solubility analysis of edible film from native and modified sago.

decline of WVTR because that film cannot absorb the water easily. WVTR of edible film greatly influenced by the properties of the polymer used, the more hydrophilic and cationic of the polymer, the higher WVTR of the film [14]. The addition of modified sago starch had improved WVTR of edible film. A similar result showed by Ulyarti *et al.* [15] that the addition of modified cassava starch decreased WVTR of film. According to Yi *et al.* [16], the decrease of WVTR may due to the increase of starch volume ratio in the film surface so that the film compactness is also increased. In this study, sample B2 has the lowest WVTR than others, which is 0,02986 g s^{-1} m^{-2} (Figure 4).

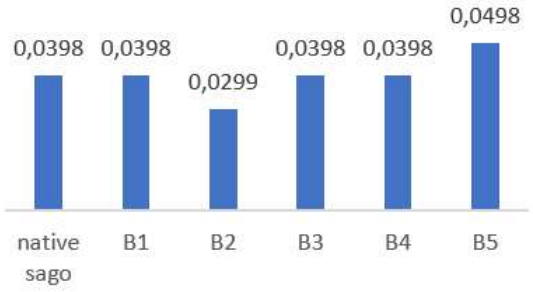


Figure 4. WVTR analysis of edible film from native and

Water solubility analysis was performed to determine the film solubility in the water. Solubility is closely related to hydrophobicity. The more hydrophobic, the lower water solubility. The addition glycerol in this formula causes the edible film more hydrophile. However, because there is no difference amount of glycerol was added to the sample, the effect of adding glycerol to each sample is considered the same. In this research, similar to moisture content and WVTR test results, edible films obtained from modified sago starch (except B5) have lower water solubility than edible films from native sago. Figure 5 showed that B2 has the lowest water solubility than others, which is 0,3352 %.

The mechanical analysis performed in this research is thickness, tensile strength, and elongation. Thickness is an important parameter because it can affect the tensile strength and percent elongation of a film. It is also affecting the type of product that can be packaged [17]. In this study, B2 and B3 have the lowest thickness, which is 0,1133 mm while B4 has the highest thickness, which is 0,17 mm (Table 3). According to the Japanese Industrial Standard (JIS), the maximum thickness of the edible film is 0,25 mm [17], so all of the edible films in this research is quite good because the thickness of it is below the standard.

Table 3. Mechanical properties of edible films.

Sample Code	Thickness (mm)	Tensile strength (N/mm^2)	Max strain (%)
native sago	0.1367± 0.01	3.8907 ± 0.03	858.9250 ±63.18
B1	0.1633 ± 0.02	4.9602 ± 0.08	37.1766 ± 7.17
B2	0.1133 ± 0.02	6.8843 ± 0.16	49.5081 ± 1.75
B3	0.1133 ± 0.00	5.3837 ± 0.05	37.7591 ± 1.42
B4	0.1700 ± 0.00	4.4258 ± 0.08	26.9104 ± 0.08
B5	0.1333 ± 0.00	4.0083 ± 0.06	34.1817 ± 0.71

The tensile test aims to determine the tensile strength of the edible film when it is under load, while the elongation test aims to determine the elongation (elasticity) of a film. The film will be stronger when the tensile test value was higher. The film also will be more elastic when the percent of elongation was higher.

Edible films made from modified sago have a higher tensile strength and smaller percent elongation compared to the edible film made from native sago. This shows that the utility of modified sago can be increasing the tensile strength and decreasing the flexibility of the edible film. A similar result showed by Ulyarti *et al.* [15] that the addition of modified cassava starch increased the tensile strength of the film. The result from Sondari *et al.* [8] also showed that biofilm made from modified sago starch using the ethanol precipitation method has higher tensile strength and percent elongation than biofilm made from native sago.

Table 3 showed that all of the edible films obtained from modified sago starch have a higher tensile strength and lower percent elongation than native sago. This result was similar to Tabari [4], he showed that the addition of CMC on the preparation of edible film based on sago starch can be decreased the elongation percent. However, B2 has the highest percent elongation than other edible films obtained from modified sago.

Three factors affect the tensile strength and percent elongation, which are intermolecular bonds (hydrogen bond, dipole-dipole, induced dipole, and London force), amylose, and amylopectin content [18]. The existence of intermolecular bonds will enhance the tensile strength of a film. Intermolecular bonds (except London Force, will also enhance the elasticity of the film therefore film obtained is not brittle. Amylose content will affect the hardness or strength of film while the amylopectin content will affect the elasticity of the film.

According to Winarti et al., [19], modified sago using butanol precipitation will enhance the amylose content and reduce the amylopectin content therefore the crystallinity was higher. However, edible films made from modified sago have a higher tensile strength and lower percent elongation than native sago.

Base on the Japanese Industrial Standard (JIS), the minimum standard of the tensile strength value of the edible film is 3,92 MPa while for the percent elongation, it will be categorized as not good if it is less than 10 % and categorized as very good if more than 50% [17]. Therefore, all of the edible films in this study have met the minimum standard of the tensile strength value. While for the percent elongation, edible films made from native sago is categorized as very good, and edible films made from modified sago is categorized as a good edible film.

CONCLUSION

The modification process using butanol precipitation method can affect the particle size, amylose, and amylopectin content of sago starch. The addition of modified sago in the preparation of edible film was able to enhance the hydrophobicity and mechanical properties of edible film. The edible films obtained from modified sago (B1, B2, B3, and B4) have better physical and mechanical properties than the edible film from native sago. The best edible film is B2 which has high tensile strength, elongation, and hydrophobicity (high contact angle, low moisture content, WVTR, and water solubility).

ACKNOWLEDGMENT

The authors would like to thank for the Ministry of Research, Technology, and Higher Education for Program Insentif Riset Sistem Inovasi Nasional (INSINAS) 2018 and the Integrated Laboratory of Bioproduct (iLab), Research Center for Biomaterial, Indonesian Institute of Science.

REFERENCES

[1] J. A. Aguirre-joya *et al.*, *Chapter 1 - Basic and Applied Concepts of Edible Packaging for Foods*. Elsevier Inc., 2017.

[2] H. Molavi, S. Behfar, M. A. Shariati, M. Kaviani, and S. Atarod, "A Review on Biodegradable Starch Based Film," *J. Microbiol. Biotechnol. Food Sci.*, vol. 4, no. 5, pp. 456–461, 2015, doi: 10.15414/jmbfs.2015.4.5.456-461.

[3] U. Shah, F. Naqash, A. Gani, and F. A. Masoodi, "Art and Science behind Modified Starch Edible Films and Coatings/ : A Review," *Compr. Rev. food Sci. food Saf.*, vol. 15, pp. 568–580, 2016, doi: 10.1111/1541-4337.12197.

[4] M. Tabari, "Characterization of a new biodegradable edible film based on Sago Starch loaded with Carboxymethyl Cellulose nanoparticles," *Nanomed Res J*, vol. 3, no. 1, pp. 25–30, 2018, doi: 10.22034/nmrj.2018.01.004.

[5] L. SW, P. D, Nurazizah, I. Z, and Bahruddin, "Bioplastic Properties of Sago-PVA Starch with Glycerol and Sorbitol Bioplastic Properties of Sago-PVA Starch with Glycerol and Sorbitol Plasticizers," *J. Phys. Conf. Ser.*, vol. 1351, 2019, doi: 10.1088/1742-6596/1351/1/012102.

[6] M. Escamilla-Garcia *et al.*, "Modified Starch-Chitosan Edible Films/ : Physicochemical and Mechanical Characterization," *Coatings*, vol. 7, no. 224, pp. 1–15, 2017, doi: 10.3390/coatings7120224.

[7] Y. Wang, R. Zhang, S. Ahmed, W. Qin, and Y. Liu, "Preparation and Characterization of Corn Starch Bio-Active Edible Packaging Films Based on Zein Incorporated with Orange-Peel Oil," *Antioxidants*, vol. 8, no. 391, pp. 1–16, 2019.

[8] D. Sondari *et al.*, "Biofilm Based on Modified Sago Starch: Preparation and Characterization," *Reaktor*, vol. 19, no. 3, pp. 125–130, 2019.

[9] Yuniar, E. Margaretty, Fadarina, M. Anerasari, and I. Febriana, "The Effect of Reaction Time and pH on the Process of Sago Starch," *IOP Conf. Ser. J. Phys.*, 2019, doi: 10.1088/1742-6596/1167/1/012053.

[10] S. H. Othman, S. A. M. Edwal, N. P. Risyon, R. K. Basha, and R. A. Talib, "Water sorption and water permeability properties of edible film made from potato peel waste," *Food Sci. Technol.*, vol. 37, no. 1, pp. 63–70, 2017.

[11] C. Dularia, A. Sinhmar, R. Thory, A. K. Pathera, and V. Nain, "International Journal of Biological Macromolecules Development of starch nanoparticles based composite fi lms from non-conventional source - Water chestnut (Trapa bispinosa)," *Int. J. Biol. Macromol.*, vol. 136, pp. 1161–1168, 2019, doi: 10.1016/j.ijbiomac.2019.06.169.

[12] C. Togas, S. Berhimpon, R. I. Montolalu, Henny Adeleida Dien, and F. Mentang, "Karakteristik Fisik Edible Film Komposit Karaginan dan Lilin Lebah Menggunakan Proses Nanoemulsi," *JPHPI*, vol. 20, no. 3, pp. 468–477, 2017.

[13] F. M. Pelissari, M. M. Andrade-mahecha, P. J. do A. Sobral, and F. C. Menegalli, "Food Hydrocolloids

Comparative study on the properties of flour and starch films of plantain bananas (Musa paradisiaca)," *Food Hydrocoll.*, vol. 30, no. 2, pp. 681–690, 2013, doi: 10.1016/j.foodhyd.2012.08.007.

[14] P. Y. Soo and N. M. Sarbon, "Preparation and characterization of edible chicken skin gelatin film incorporated with rice flour," *Food Packag. Shelf Life*, vol. 15, no. August 2017, pp. 1–8, 2018, doi: 10.1016/j.fpsl.2017.12.009.

[15] U. Ulyarti, N. Nazarudin, Surhaini, Lisani, R. Ramadon, and P. Lumbanraja, "Cassava Starch Edible Film with Addition of Gelatin or Modified Cassava Starch Cassava Starch Edible Film with Addition of Gelatin or Modified Cassava Starch," *IOP Conf. Ser. Earth Enviromental Sci.*, vol. 515, 2020, doi: 10.1088/1755-1315/515/1/012030.

[16] J. B. Yi, Y. T. Kim, H. J. Bae, W. S. Whiteside, and H. J. Park, "Influence of Transglutaminase-Induced Cross-Linking on Properties of Fish Gelatin Films," *Jpurnal Food Sci.*, vol. 71, no. 9, pp. 376–383, 2006, doi: 10.1111/j.1750-3841.2006.00191.x.

[17] S. Khairunnisa, Juniarto, Zahidah, and I. Rostini, "The effect of glycerol concentration as a plasticizer on edible films made from alginate towards its physical characteristic," *World Sci. News*, vol. 112, no. October, pp. 130–141, 2018.

[18] Y. E. Agustin and K. S. Padmawijaya, "Sintesis Bioplastik dari Kitosan-Pati Kulit Pisang Kepok dengan Penambahan Zat Aditif," *J. Tek. Kim.*, vol. 10, no. 2, pp. 40–48, 2016.

[19] C. Winarti, N. Richana, D. Mangunwidjaja, and T. . Sunarti, "Effect of Arrowroot Nano Starch Preparation Methods on the Characteristics of Temulawak Oleoresin Microcapsules Effect of Arrowroot Nano Starch Preparation Methods on the Characteristics of Temulawak Oleoresin Microcapsules," *IOP Conf. Ser. Earth Enviromental Sci.*, vol. 309, pp. 1–7, 2019, doi: 10.1088/1755-1315/309/1/012034.

Copyright © 2019 Jusami | Indonesian Journal of Materials Science. This article is an open access article distributed under the terms and conditions of the Creative Commons Attribution-NonCommercial-ShareAlike 4.0 International License (CC BY-NC-SA 4.0).