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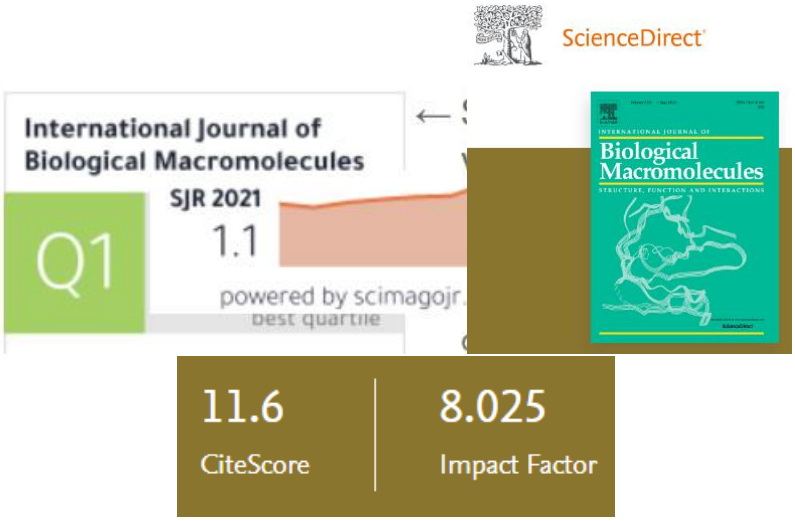

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Characterization and Properties of Cellulose Microfibers from Water Hyacinth Filled Sago Starch Biocomposites

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Characterization and Properties of Cellulose Microfibers from Water Hyacinth Filled Sago Starch Biocomposites

The cellulose microfibers (CMF) from water hyacinth (WH) fiber as a filler in sago starch (SS) biocomposites was investigated. The CMF was isolated by pulping, bleaching and acid hydrolysis methods. The addition of CMF in sago matrix was varied i.e. 0, 5, 10, 15 and 20 wt%. Biocomposites were made by using solution casting and glycerol as a plasticizer. The biocomposites were also determined by tensile test, FTIR, X-Ray, thermogravimetric, SEM, and soil burial tests. The results show that the SS15CMF sample has the highest tensile strength of 10.23 MPa than those other samples. Scanning Electron Microscope (SEM) images show that the strong interaction was formed between CMF WH and matrix. Fourier Transform Infra-red (FTIR) indicated that the functional group of biocomposites was a hydrophilic cluster. The addition of CMF WH in sago starch biocomposites lead to the moisture barrier, crystallinity, and thermal stability increased; it is due to the pure sago starch film was more rapidly degraded than its biocomposites.

Keywords: water hyacinth fiber, cellulose microfibers, sago starch, natural fiber, biocomposites, bioplastics

1. Introduction

Synthetic plastic become serious problem in every country due to its not environmentally friendly and resulting in waste pollution. These cases were released Jambeck et al. [1] that Indonesia is the biggest country which produce plastic in ocean after China [1] (Jambeck et al., 2015). This is an emergency phenomenon due to its effect in environment and society. Thus, many researchers interest to develop the manufacturing bioplastic from starch in one last decade (Asrofi et al., 2018).

Starch is a kind of biopolymer which can be obtained by extraction from vegetable ingredients containing carbohydrates such as tuber and cereal. Manufacturing starch-based bioplastic has been got a lot of attention due to its biodegradability, low cost and abundant in earth [2] (López et al., 2011). The finding of biodegradable material is needed to overcome the waste pollution of synthetic plastic. Two last decades, many researchers focused on natural fiber-reinforced polymer composites rather than synthetic fiber-reinforced composites [3,4] (Wahono et al., 2018; Senthamaraikannan, et al., 2016). Most of polymer researchers still use synthetic resin as a composite polymer matrix. The disadvantage of composite from resin matrix is only partial degradation in environment. Therefore, to make fully biodegradable materials, we have to use biodegradable matrix derived from natural materials as well. Sago starch is one of the material candidates for biodegradable matrix due to its commercially available throughout the world [5] (Tongdeesoontorn et al., 2011).

However, sago starch is brittle and resulting in poor mechanical properties. To overcome this weakness, plasticizer is added to starch which can reduce the fragility of the material and improve the capability process [6] (Chang et al., 2006). Apart from this, starch has a higher moisture absorption and low thermal stability properties. The addition of cellulose fibers in starch matrix is one of the efficient methods to improve the performance of starch-based composites. The hydrophilic character of cellulose fiber and starch can produce a good

hydrogen bonding between the two. Fiber size also plays an important role in properties of biocomposites [7,8] (Asrofi et al., 2018; Sanjay et al. 2018). The smaller fiber size then resulting in higher contact surface area [9] (Abral, Lawrensius, Handayani, &Sugiarti, 2018a).

Nowdays, biocomposite based starch matrix have used as an alternative food packaging in substituting the synthetic plastic [10,11] (Sumrith et al., 2019; Marichelvam et al., 2019). These previous studied have reported that the main advantage of starch matrix is low cost, abundant and biodegradability in environment [12,3] (Sanjay et al., 2016; Wahono, Irwan, Syafri, &Asrofi, 2018). The tensile strength, thermal stability and moisture barrier of biocomposite to increased [13,14] (Lopez et al., 2015: Asem, Nawawi, & Jimat, 2017).

Furthermore, several studies reported about the addition of cellulose fiber in microscale reinforced starch matrix. Various sources of cellulose fiber were used to filled starch matrix biocomposites such as oil empty fruit bunch (Abral et al., Putra, Asrofi, Park, & Kim, 2018b), okara fiber (Fu et al., 2017), micro scale pulp water hyacinth [15-20] (Abral at al., 2018; (Asrofi, Abral, Putra, Sapuan, & Kim, 2018). They just given pulping treatment in resulting micro cellulose fiber. This treatment is considered to be insignificant to increase the surface adhesion between fibers and matrix. Therefore, in this study, we have isolated the CMF WH by pulping, bleaching and hydrolysis treatment. According to previous report, three stage treatment above give several advantages such as higher crystallinity and cellulose content [21, **7/17/18/19**] (**Asrofi et al., 2018**; Manimaranet al., 2018). Then, CMF WH was prepared and filled in sago starch matrix to produce biocomposites. They were characterized through SEM, tensile test, X-ray diffraction (XRD), FTIR, (Thermogravimetric Analysis) TGA, moisture absorption and soil burial test.

2. Materials and Methods

2.1. Materials

Water hyacinth fiber was obtained from Payakumbuh, Indonesia. The isolation of MFC WH fiber was explained in methods. Sago starch was purchased at local market in Padang, Indonesia. Glycerolunder Brataco brand with density 1.255–1.260 g/mL was purchased from PT Cisadane Raya Chemicals, Tangerang Indonesia. All chemical reagent (pure analyst) for CMF isolation such as NaOH, KOH, $NaClO_2$, acetic acid and H_2SO_4, were supplied from Faculty of Agriculture Technology, Andalas University, Indonesia.

2.2. Preparation Isolation of CMF WH

The isolation of CMF WH can be seen in Fig. 1. The WH stem was separated from leaves and roots. Then, it was cut along 10-20 mm and dried under the sun for 3 days with water content 9-10% [**7/17/18/19**] (Asrofi et al., 2018). The lignin and hemicellulose of WH fiber were removed by pulping digester (simple digester pulp) in 18% NaOH solution at 170 ^oC and 7-9 kg/cm^2 for 2 h [22] (Syafri, Kasim, Abral,&Asben, 2018). The pulped WH fiber was rinsed by distilled water until free from alkali and continued by bleaching process with 5% $NaClO_2$: acetic acid at 70 ^oC for 2 h. After that, the bleached WH fiber was neutralized then it was hydrolyzed by 30% H_2SO_4 for 30 min. The final product of CMF WH was in dried granule fiber as in Fig. 1e.

2.3. Fabrication of CMF WH/SS Biocomposites

The fabrication of biocomposites referred to previous report [23] (Syafri et al., 2018). SS was dissolved by aquadest (5% w/v) and CMF WH (% w/w from SS) with different composition in matrix (see Table 1). Glycerol (30% w/w from SS) was added as a plasticizer. Then, they were mixed become biocomposite solution at 90 ^oC under constant stirring (300 rpm) until gelatinized. The biocomposites gel was casted in rectangular acrylic mold

(dimension: 11 x 9.5 x 0.3 cm3). To remove an air bubble in biocomposites gel, the rectangular acrylic mold was placed in ultrasonic bath for 15 min. After that, it was dried in drying oven at 40 ^{o}C for 24 h. The biocomposites film was stored in desiccator for 24 h before characterization.

2.4. Fracture Surface

The fracture surface of all samples after tensile test were observed by SEM Hitachi 3400 Seri N. The voltage was operated at 10 kV. The test was carried out in room temperature.

2.5. Tensile Test

The tensile test of all biocomposites sample was determined by UTM Strograph-R1 instrument with load cell 5 kN. All samples were prepared and formed according to American Standard Testing Material (ASTM) D-638. The crosshead speed and temperature during test were monitored at 2 mm/min and 25 ^{o}C, respectively. The tensile test was carried out by three times repetition for each sample variation [24,25] (Sari, Wardana, Irawan, & Siswanto, 2017: Sari, et al., 2019).

2.6. Crystallinity Index

The crystallinity index (CI) of biocomposites was studied by X-Ray Diffraction (XRD) PANalytical. The XRD profile was recorded by diffractometer ray circuit. The radiation was CuKα at wave length 1.5406Å. The ampere and voltage were operated at 30 mA and 40 kV, respectively. The 2θ degree was scanned in range 2 – 100^{o} every 20 ^{o}/min. The CI (%) was calculated by Segal's equation [26] (Segal, Creely, Martin, & Conrad, 1959):

$$CI\ (\%) = \frac{I_c - I_{am}}{I_c} x100 \qquad (1)$$

Where, I_c and I_{am} were crystalline and amorphous region, respectively. I_c was measured at 2θ = 22.6^{o}. Meanwhile, I_{am} was determined at 2θ = 18^{o}.

2.7. Functional Groups

The functional group of all sample tested was determined by FTIR using Perkin-Elmer Frontier. All samples were formed in square film 1 cm x 1 cm. The spectrum was scanned from wavenumber 4000-600 cm^{-1} under resolution 4 cm^{-1}.

2.8. Thermal Stability

Thermogravimetric analysis was conducted by thermogravimetric analyzer instrument. All samples were tested under nitrogen atmosphere as heat source with 80 ml/min of speed. The test was done in range temperature 30 – 600 ^{o}C. The heating rate was 20 ^{o}C /min.

2.9. Moisture Absorption Test

The biocomposites sample was cut 1.5 cm × 1.5 cm and dried until constant weight in oven at 40 ^{o}C for 24 h. The test was done in moisture container at relative humidity and temperature of 80% and 25 ^{o}C for 22 h, respectively. W_o and W_t were initial weight before test and final weight of sample after measured in 30 min intervals, respectively. The percentage of moisture absorption was calculated according to equation below:

$$Moisture\ Absorption\ (\%) = \frac{W_t - W_0}{W_0} x100 \quad (2)$$

2.10 Biodegradation Test

Soil burial test method was used to determine the biodegradation of biocomposites sample [27] (Pimpan, Ratanarat, & Pongchawanakul, 2001). The studied soil was the community plantation soil in Padang. The pH water content and relative humidity were 6.5, 36.24% and 78%, respectively. All samples were dried in drying oven at 40 ^{o}C for 24 h until constant weight and then weighted for initial weight with precision balance. The samples were buried in soil for 5 and 15 days. After that, they were cleaned with distilled water and dried in oven at 40 ^{o}C for 24 h. Then, they were stored in desiccator for 24 h before final weighing. The

final weighing was done by precision balance until constant weight. The percentage weight loss of all samples was determined according to equation below:

$$Weight\ loss\ (\%) = \frac{W_0 - W_t}{W_t} x100 \quad (3)$$

Where, W_o and W_t were the sample weight before and after burial in soil, respectively.

3. Result and Discussions

3.1. Chemical composition of WH Fiber

Chemical composition analysis was used to determine the percentage of cellulose, hemicellulose, lignin and extractive content in WH fiber. The chemical composition of WH fiber before and after chemical treatment is presented on Table 2. The untreated WH fiber (raw) has cellulose, hemicellulose, lignin and extractive content for 64.07, 15.13, 10.48 and 5.26 wt%, respectively. The cellulose content in raw WH fiber is lower than treated fiber. Giving chemical treatment is one of the ways to reduce the non-cellulosic content and increase the cellulose content. It can be seen that lignin and hemicellulose content decreased after pulping process with NaOH. It is due to the broken of hydrogen bond in cell wall structure of fiber [28] (Chandra, George, &Narayanankutty, 2016).

Meanwhile, there is a significant improvement of cellulose content after bleaching and hydrolysis process (Table 2). The cellulose content increased 14% from pulping to bleaching process due to the removal of hemicellulose and lignin content in WH fiber. Lignin reacted with $NaClO_2$ and resulting in soluble lignin chloride compound [29] (Chirayil et al., 2014). After hydrolysis process, the cellulose content of CMF WH fiber has 85.85%. This value is higher than previous study which reported about WH cellulose fiber prepared by double acid hydrolysis [**7/17/18/19**] (Asrofi et al.,2018).

3.2. Fracture Morphological of Biocomposites

Fracture morphology of biocomposites after tensile test is displayed by Fig. 2. Generally, the morphological of biocomposite without filler (SS film) is uniform, homogeneous and no porosity formation [30] (Kargarzadeh, Johar, & Ahmad, 2017). The addition of 5 and 10 wt%CMF WH in SS matrix shows an agglomeration fiber and porosity (Fig. 2b and 2c). It is indicated that in several fracture point occurs a bad adhesion bonding between cellulose fiber and matrix. However, the presence of cellulose fiber in starch matrix improved the mechanical properties of biocomposites than SS film. Fig.2d displays non-homogeneous distribution of cellulose fiber in matrix due to unperfect mixing of biocomposites during fabrication. According to the previous report, unperfect mixing able to provoke the agglomeration, porosity and bad adhesion bonding [**7/17/18/19**] (Asrofi et al., 2018). This phenomenon is affected in decreasing tensile test.

3.3. Tensile Strength

The tensile strength and strain at break of SS film and its biocompositesare showed by Fig. 3. The tensile strength increased with the addition of CMF WH in SS matrix (Fig. 3a). It can be seen that the tensile strength of SS film is 3.77 MPa. It is lower than its biocomposites due to there is no reinforcement in this sample. After addition as much 5, 10 and 15wt%, the tensile strength increased significantly become 7.12, 9.58 and 10.23 MPa, respectively. This improvement is followed by decreasing of strain value (Fig. 3b). This is due to a good interaction between cellulose fiber and starch [31] (Lendvai, Karger Kocsis, Kmetty, & Drakopoulos, 2016). Another phenomenon occurs in SS20CMFWH sample. There is a decreasing in tensile strength from 10.23 to 10.06 MPa. This phenomenon due to inhomogeneous distribution of fiber in starch matrix [32] (Savadekar&Mhaske, 2012). This case is agreement with SEM and XRD observation.

3.4. Crystallinity Index Analysis

Fig. 4 shows diffraction pattern of SS film and its biocomposites. It can be seen that all biocomposites have same pattern and the crystalline area appeared with the addition of CMF WH in SS matrix. It is indicated that biocomposites material included in semi-crystalline structure [30] (Kargarzadeh, Johar, & Ahmad, 2017). At Fig. 4, SS films did not show sharp peaks. It is indicated due to there is no presence of cellulose fiber in matrix. Different phenomena are shown by biocomposites sample. There are several peaks in the range $2\theta = 16 - 24^{o}$ especially for SS15CMFWH and SS20CMFWH sample. The CI of SS film, SS5CMFWH, SS10CMFWH SS15CMFWH, SS20CMFWH was 14.01, 19.48, 19.95, 40.72, 33.98%, respectively. This case proves that the presence of cellulose fiber in matrix give a rise the peaks and increasing CI. It can be seen that the highest CI is owned by SS15CMFWH and SS20CMFWH sample. According to previous research, increasing CI also trigger an increase in tensile strength [**7/17/18/19**] (Asrofi et al., 2018). This is indicated by the presence of cellulose fiber chains inhibit the movement of starch polymers and resulting in brittle biocomposites [33] (Sanyang, Sapuan, Jawaid, Ishak, &Sahari, 2016). This result is supported by the lower strain value with increasing cellulose fraction in matrix (Fig. 3).

3.5. Functional Group Analysis

The chemical functional groups in CMF WH filled sago starch matrix based biocomposites are presented in Fig. 5. IR spectra of all samples had similar bands. There is no significant different between SS film and its biocomposites. This phenomenon indicated that the addition of glycerol or CMF WHF in sago matrix is not affect the wavenumber shift [**22/23**] (Syafri et al., 2018).

There are five different bands exist in all biocomposites sample i.e. 3298, 2926, 1647, 1353 and 1009 cm^{-1}. The bands around 3298 $cm^{-1}$is indicated by OH stretching groups from SS and CMF WH due to the presence of hydroxyl groups in in both of these materials [34,

7/17/18/19] (Kaewtatip & Thongmee, 2012; Asrofi et al., 2018). The vibration stretching CH occurred in band around 2926 cm^{-1}. Meanwhile, the band around 1647 cm^{-1} indicated by the OH water absorption of natural hydrophilic starch and cellulose [**9/15/16**] (Abral et al, 2018). This band appear in all sample tested. The band at 1353 cm^{-1} and 1647 cm^{-1}indicated of a strong and broad of CO stretch which is a cellulose alcohol group [35] (Mahardika, Abral, Kasim, Arief, &Asrofi, 2018).

3.6. Thermal Stability

Thermal analysis of CMF WH filled starch based bioocompositeis shown in Fig. 6. There are three stage degradation temperature. The first stage occurs below 100 ^oC due to moisture loss in all biocomposites sample [36] (Lee et. al., 2009). At the second stage, the degradation temperature starts from 225 until 350^oC. During this process, the ether bonds and unsaturated structures occur condensation between hydroxyl groups of starch chains. In this stage, starch, cellulose and glycerol structure were all degraded [**7/17/18/19]** (Asrofi et al., 2018). The last stage occurs at temperatures above 350 ^oC due to the residual disintegration produced in the previous conditions [37] (Raabe et al., 2015). From all stage, we can conclude that the highest thermal stability was inSS20CMFWH sample with a remaining mass of 30.6% (Fig. 6a). This result is supported by DTG curve (Fig. 6b).

It can be seen that the SS20CMFWH sample has the highest thermal degradation of 332.71 ^oC with a thermal degradation rate of 0.95 %/min. It is due to good interaction of hydrogen bonding between CMF and SS matrix [30] (Kargarzadeh, Johar, & Ahmad, 2017). This phenomenon was contradictive with SS film. SS film has the lowest thermal stability. Its degradation temperature is about 328.08$^oC$with a degradation rate of 1.43%/min. This case is similar with previous report [38] (Zainuddin, Ahmad, Kargarzadeh, Abdullah, & Dufresne, 2013). Previous study reported about the addition of fiber into starch matrix was successful improved thermal stability. This is due to good adhesion bonding between fiber and starch

which resulting in little of sample weight loss [39] (Prachayawarakorn, Chaiwatyothin, Mueangta, &Hanchana, 2013).

3.7. Moisture Absorption

The disadvantage of cellulose and starch-based biocomposites is the high absorption of moisture due to its hydrophilic character [**9/15/16**] (Abral et al., 2018). Fig. 7 displays the percentage of moisture absorption for 22 h. It can be seen that there is a significant different between SS film and its biocomposites. At the beginning, the sample absorbs a moisture in high capacity. The moisture absorption percentage of SS Films, SS5CMFWH, SS10CMFWH, SS15CMFWH and SS20CMFWH at 3.5 h is 28.58, 24.55, 24.71, 23.37 and 23.01%, respectively. Moisture absorption rate decrease towards to the saturation point at 5.5 h.

It can be seen that addition of CMF WH in SS matrix reduce the moisture absorption. The lowest moisture absorption was in SS20CMFWH samples. This is due to CMF is an organic substance has less hydrophilic than SS. Another reason was indicated the role of CMF WH as barrier agent of water molecule when diffusing in to matrix [**22/23**,**7/17/18/19]**] (Syafri et al., 2018; Asrofi et al., 2018).

3.8. Soil Burial Test

Biodegradation testing of biocomposites was carried out by buring the sample in the soil. The percentage weight loss due to biodegradation is presented in Fig. 8. The highest percentage of weight loss was in SSFilm and SS5CMFWH after 15 days. This phenomenon is indicated by the absence of fibers in SS matrix. This sample easily broken down by microorganisms. Another case occured in SS20CMFWH sample where it has the lowest weight loss. This is due to the role of CMF WH in SS matrix as a barrier agent to prevent microorganisms. Previous research also showed the similar case that the addition of

montmorillonite in poly (butylene succinate) matrix reduced the biodegradation rate [40] (Phua, Lau, Sudesh, Chow, & Ishak, 2012).

4. Conclusions

The biocomposites based CMF WH and SS matrix was successfully produced through solution casting method. The presence of CMF WH in SS improved tensile strength, thermal stability and CI. The maximum tensile strength was 10.23 MPa achieved by SS15CMFWH sample. There is no significant change in functional group with the addition of fiber in matrix. The lowest moisture absorption was in SS20CMFWH for 23.01%. The addition of fiber in matrix also reduced the biodegradation rate in soil. This biocompositesis suggested for food packaging application due to its excellent properties and environmentally friendly.

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References

[9] Abral, H., Lawrensius, V., Handayani, D., &Sugiarti, E. (2018a). Preparation of nano-sized particles from bacterial cellulose using ultrasonication and their characterization. *Carbohydrate Polymers*, *191*, 161-167.

[16] Abral, H., Putra, G. J., Asrofi, M., Park, J. W., & Kim, H. J. (2018). Effect of vibration duration of high ultrasound applied to bio-composite while gelatinized on its properties. *Ultrasonics sonochemistry*, *40*, 697-702.

[15] Abral, H., Dalimunthe, M. H., Hartono, J., Efendi, R. P., Asrofi, M., Sugiarti, E.,Sapuan, S. M., Park, J. W., & Kim, H. J. (2018). Characterization of tapioca starch biopolymer composites reinforced with micro scale water hyacinth fibers. *Starch-Stärke*, *70*, 1-8.

[14] Asem, M., Nawawi, W. M. F. W., & Jimat, D. N. (2018). Evaluation of water absorption of polyvinyl alcohol-starch biocomposite reinforced with sugarcane bagasse nanofibre: Optimization using Two-Level Factorial Design. *IOP Conference Series: Materials Science and Engineering*, *368*, 012005.

[17] Asrofi, M., Abral, H., Kasim, A., Pratoto, A., Mahardika, M., &Hafizulhaq, F. (2018). Characterization of the sonicated yam bean starch bionanocomposites reinforced by nanocellulose water hyacinth fiber (WHF): The effect of various fiber loading. *Journal of Engineering Science and Technology*, *13*, 2700-2715.

[7] Asrofi, M., Abral, H., Kasim, A., Pratoto, A., Mahardika, M., &Hafizulhaq, F. (2018). Mechanical properties of a water hyacinth nanofiber cellulose reinforced thermoplastic starch bionanocomposite: Effect of ultrasonic vibration during processing. *Fibers*, *6*, 40.

[18] Asrofi, M., Abral, H., Putra, Y. K., Sapuan, S. M., & Kim, H. J. (2018). Effect of duration of sonication during gelatinization on properties of tapioca starch water hyacinth fiber biocomposite. *International Journal of Biological Macromolecules*, *108*, 167-176.

[19] Asrofi, M., Abral, H., Kasim, A., Pratoto, A., Mahardika, M., Park, J. W., & Kim, H. J. (2018). Isolation of nanocellulose from water hyacinth fiber (whf) produced via digester-sonication and its characterization. *Fibers and Polymers*, *19*, 1618-1625.

[28] Chandra, J., George, N., &Narayanankutty, S. K. (2016). Isolation and characterization of cellulose nanofibrils from arecanut husk fibre. *Carbohydrate Polymers*, *142*, 158-166.

[6] Chang, Y. P., Karim, A. A., &Seow, C. C. (2006). Interactive plasticizing–antiplasticizing effects of water and glycerol on the tensile properties of tapioca starch films. *Food Hydrocolloids*, *20*, 1-8.

[29] Chirayil, C. J., Joy, J., Mathew, L., Mozetic, M., Koetz, J., & Thomas, S. (2014). Isolation and characterization of cellulose nanofibrils from Helicteresisora plant. *Industrial Crops and Products*, *59*, 27-34.

[20] Fu, Z. Q., Wu, M., Han, X. Y., & Xu, L. (2017). Effect of okara dietary fiber on the properties of starch-based films. *Starch-Stärke*, *69*, 1700053.

[1] Jambeck, J. R., Geyer, R., Wilcox, C., Siegler, T. R., Perryman, M., Andrady, A., & Law, K. L. (2015). Plastic waste inputs from land into the ocean. *Science*, *347*, 768-771.

[34] Kaewtatip, K., &Thongmee, J. (2012). Studies on the structure and properties of thermoplastic starch/luffa fiber composites. *Materials & Design*, *40*, 314-318.

[30] Kargarzadeh, H., Johar, N., & Ahmad, I. (2017). Starch biocomposite film reinforced by multiscale rice husk fiber. *Composites Science and Technology*, *151*, 147-155

[36] Lee, S. Y., Mohan, D. J., Kang, I. A., Doh, G. H., Lee, S., & Han, S. O. (2009). Nanocellulose reinforced PVA composite films: effects of acid treatment and filler loading. *Fibers and Polymers*, *10*, 77-82.

[31] Lendvai, L., Karger-Kocsis, J., Kmetty, Á., &Drakopoulos, S. X. (2016). Production and characterization of microfibrillated cellulose-reinforced thermoplastic starch composites. *Journal of Applied Polymer Science*, *133*, 1-8.

[2] López, O. V., Lecot, C. J., Zaritzky, N. E., & García, M. A. (2011). Biodegradable packages development from starch based heat sealable films. *Journal of Food Engineering*, *105*, 254-263.

[13] López, O. V., Castillo, L. A., García, M. A., Villar, M. A., & Barbosa, S. E. (2015). Food packaging bags based on thermoplastic corn starch reinforced with talc nanoparticles. *Food Hydrocolloids*, *43*, 18-24.

[35] Mahardika, M., Abral, H., Kasim, A., Arief, S., &Asrofi, M. (2018). Production of nanocellulose from pineapple leaf fibers via high-shear homogenization and ultrasonication. *Fibers*, *6*, 28.

[21] **Manimaran**, P., Senthamaraikannan, P., Sanjay, M. R., Marichelvam, M. K., &Jawaid, M. (2018). Study on characterization of furcraeafoetida new natural fiber as composite reinforcement for lightweight applications. *Carbohydrate Polymers*, *181*, 650-658.

[21] **Manimaranet?**

[11] Marichelvam, M. K., Jawaid, M., & Asim, M. (2019). Corn and Rice Starch-Based Bio-Plastics as Alternative Packaging Materials. *Fibers*, *7*, 32.

[27] Pimpan, V., Ratanarat, K., &Pongchawanakul, M. (2001). Preliminary study on preparation of biodegradable plastic from modified cassava starch. *Journal of Scientific Research Chulalongkorn University*, *26*, 117-126.

[39] Prachayawarakorn, J., Chaiwatyothin, S., Mueangta, S., &Hanchana, A. (2013). Effect of jute and kapok fibers on properties of thermoplastic cassava starch composites. *Materials & Design*, *47*, 309-315.

[40] Phua, Y. J., Lau, N. S., Sudesh, K., Chow, W. S., & Ishak, Z. M. (2012). Biodegradability studies of poly (butylene succinate)/organo-montmorillonite nanocomposites under controlled compost soil conditions: effects of clay loading and compatibiliser. *Polymer Degradation and Stability*, *97*, 1345-1354.

[37] Raabe, J., Fonseca, A. D. S., Bufalino, L., Ribeiro, C., Martins, M. A., Marconcini, J. M.,&Tonoli, G. H. D. (2015). Biocomposite of cassava starch reinforced with cellulose

pulp fibers modified with deposition of silica (SiO2) nanoparticles. *Journal of Nanomaterials*, *2015*, 1-9.

[12] Sanjay, M. R., Arpitha, G. R., Naik, L. L., Gopalakrishna, K., & Yogesha, B. (2016). Applications of natural fibers and its composites: An overview. *Natural Resources*, *7*, 108.

[8] Sanjay, M. R., Madhu, P., Jawaid, M., Senthamaraikannan, P., Senthil, S., & Pradeep, S. (2018). Characterization and properties of natural fiber polymer composites: A comprehensive review. *Journal of Cleaner Production*, *172*, 566-581.

[33] Sanyang, M. L., Sapuan, S. M., Jawaid, M., Ishak, M. R., &Sahari, J. (2016). Effect of sugar palm-derived cellulose reinforcement on the mechanical and water barrier properties of sugar palm starch biocomposite films. *BioResources*, *11*, 4134-4145.

[25] Sari, N. H., Wardana, I. N., Irawan, Y. S., &Siswanto, E. (2017). Corn Husk Fiber-Polyester Composites as Sound Absorber: Nonacoustical and Acoustical Properties. *Advances in Acoustics and Vibration*, *2017*, 1-7.

[24] Sari, N. H., Sanjay, R. M., Arpitha, G. R., Pruncu, C. I., Siengchin, S. (2019). Synthesis And properties of pandanwangi fiber reinforced polyethylene composites: Evaluation of dicumyl peroxide (DCP) effect. composites Communications, *15*, 53-57.

[32] Savadekar, N. R., &Mhaske, S. T. (2012). Synthesis of nano cellulose fibers and effect on thermoplastics starch based films. *Carbohydrate Polymers*, *89*, 146-151.

[26] Segal, L. G. J. M. A., Creely, J. J., Martin Jr, A. E., & Conrad, C. M. (1959). An empirical method for estimating the degree of crystallinity of native cellulose using the X-ray diffractometer. *Textile Research Journal*, *29*, 786-794.

[4] Senthamaraikannan, P., Saravanakumar, S. S., Arthanarieswaran, V. P., &Sugumaran, P. (2016). Physico-chemical properties of new cellulosic fibers from the bark of Acacia planifrons. *International Journal of Polymer Analysis and Characterization*, *21*, 207-213.

[10] Sumrith, N., Rangappa, S. M., Dangtungee, R., Siengchin, S., Jawaid, M., Pruncu, C. I., Biopolymers-Based Nanocomposites: Properties and Applications, Springer Cham, Bio-based Polymers and Nanocomposites, 2019, pp. 255-272.

[22] Syafri, E., Kasim, A., Abral, H., &Asben, A. (2018). Cellulose nanofibers isolation and characterization from ramie using a chemical-ultrasonic treatment. *Journal of Natural Fibers*, *2018*, 1-11.

[23] Syafri, E., Kasim, A., Abral, H., Sulungbudi, G. T., Sanjay, M. R., & Sari, N. H. (2018). Synthesis and characterization of cellulose nanofibers (CNF) ramie reinforced cassava

starch hybrid composites. *International Journal of Biological Macromolecules*, *120*, 578-586.

[5] Tongdeesoontorn, W., Mauer, L. J., Wongruong, S., Sriburi, P., &Rachtanapun, P. (2011). Effect of carboxymethyl cellulose concentration on physical properties of biodegradable cassava starch-based films. *Chemistry Central Journal*, *5*, 1-8.

[3] Wahono, S., Irwan, A., Syafri, E., &Asrofi, M. (2018). Preparation and characterization of ramie cellulose nanofibers/caco3 unsaturated polyester resin composites. *ARPN Journal of Engineering and Applied Sciences*, *13*, 746-51.

[38] Zainuddin, S. Y. Z., Ahmad, I., Kargarzadeh, H., Abdullah, I., & Dufresne, A. (2013). Potential of using multiscale kenaf fibers as reinforcing filler in cassava starch-kenaf biocomposites. *Carbohydrate Polymers*, *92*, 2299-2305.

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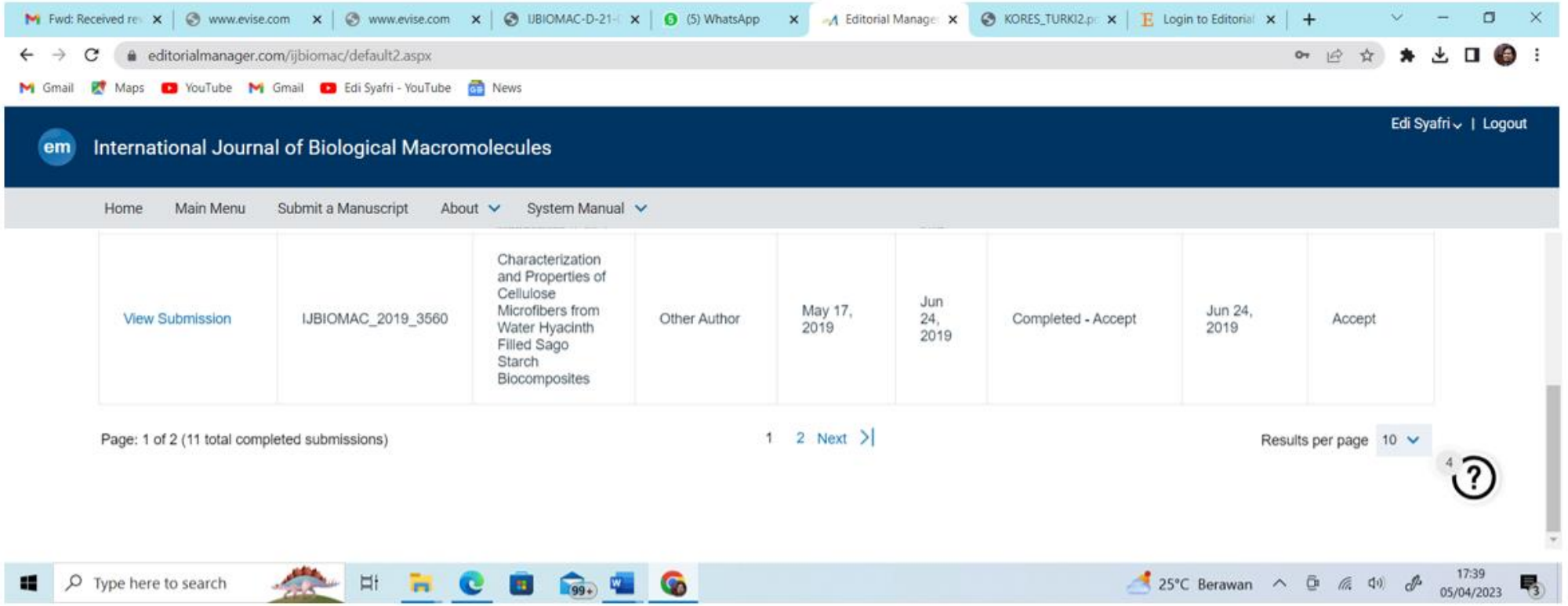
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Characterization and properties of cellulose microfibers from water hyacinth filled sago starch biocomposites

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ABSTRACT

The cellulose microfibers (CMF) from water hyacinth (WH) fiber as a filler in sago starch (SS) biocomposites was investigated. The CMF was isolated by pulping, bleaching and acid hydrolysis methods. The addition of CMF in sago matrix was varied i.e. 0, 5, 10, 15 and 20 wt%. Biocomposites were made by using solution casting and glycerol as a plasticizer. The biocomposites were also determined by tensile test, FTIR, X-Ray, thermogravimetric, SEM, and soil burial tests. The results show that the SS15CMF sample has the highest tensile strength of 10.23 MPa than those other samples. Scanning Electron Microscope (SEM) images show that the strong interaction was formed between CMF WH and matrix. Fourier Transform Infra-red (FTIR) indicated that the functional group of biocomposites was a hydrophilic cluster. The addition of CMF WH in sago starch biocomposites lead to the moisture barrier, crystallinity, and thermal stability increased; it is due to the pure sago starch film was more rapidly degraded than its biocomposites.