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Paper 4

Effect of sonication time on thermal stability, moisture absorption and biodegradation of nanocellulose water hyacinth (Eichhornia crassipes) filled bengkuang (Pachyrhizus erosus) starch biocomposites

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Short title	-
Article type	Original article

Abstract

In Indonesia, starch, especially from bengkuang (Pachyrhizus erosus), is abundant and inexpensive, thereby increasing the value of bengkuang starch through its role in mixing with bioplastic-based starch. Biocomposite from nanocellulose water hyacinth (Eichhornia crassipes) fibre and bengkuang (Pachyrhizus erosus) starch was successfully fabricated using the solution casting method. The nanocellulose content in the matrix was kept constantat 1 wt%. During fabrication, a biocomposite gel was treated by ultrasonic bath for 0, 15, 30 and 60 min.
Thermogravimetric analysis, moisture absorption, Fourier transform infrared and scanning electron microscopy were performed. The biocomposite sample with 60 min vibration has the highest thermal stability and exhibits low moistureabsorption. The soil burial test proves that this biocomposite has a slower biodegradation rate than the 0 min vibratedsample. This result was supported by morphological evaluation after biodegradation in which the 60 min vibrated sample has a coarse surface and low porosity formation.

Keywords	Nanocellulose, biodegradation, biocomposites, hyacinth fibre thermogravimetricanalysis.
Taxonomy	Elemental Analysis, Thermal Analysis, Moisture Testing

Corresponding Author	edi syafri
Order of Authors	edi syafri, Ahmad Fudholi
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Effect of sonication time on thermal stability, moisture absorption and biodegradation of nanocellulose water hyacinth (*Eichhornia crassipes*) filled bengkuang (*Pachyrhizus erosus*) starch biocomposites

Edi Syafri[1*], Sudirman[2], Mashadi[2], Evi Yulianti[2], Deswita[2], Mochamad Asrofi[3], Hairul Abral[4], R.A. Ilyas[5,6], Ahmad Fudholi[7]

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2. Revision 1 (R1)

Manuscript Details

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Short title	-
Article type	Original article

Abstract

In Indonesia, starch, particularly that obtained from bengkuang (Pachyrhizus erosus), is abundant and inexpensive, thereby increasing the value of bengkuang starch, which can be mixed with bioplastic-based starch. A biocomposite comprising nanocellulose from water hyacinth (Eichhornia crassipes) and bengkuang starch was successfully fabricated using the solution casting method. Nanocellulose content in the matrix was kept constant at 1 wt%. Moreover, during fabrication, the biocomposite gel was treated in an ultrasonic bath for 0, 15, 30, and 60 min. Further, thermogravimetric analysis, moisture absorption measurements, Fourier transform infrared spectroscopy, and scanning electron microscopy were performed. The biocomposite sample vibrated for 60 min had the highest thermal stability and exhibited low moisture absorption. The soil burial test proved that this biocomposite, as opposed to 0-min vibrated samples, has a slower biodegradation rate. This result was supported by morphological evaluation after biodegradation, in which the 60-min vibrated samples showed a coarse surface and low porosity formation.

Keywords	Nanocellulose, biodegradation, biocomposites, hyacinth fiber, thermogravimetric analysis
Taxonomy	Elemental Analysis, Thermal Analysis, Moisture Testing

Corresponding Author	edi syafri
Order of Authors	edi syafri, Sudirman Sudirman, Mashadi Mashadi, Evi Yulianti, Deswita Deswita, Mochamad Asrofi, Hairul Abral, R.A. Ilyas, S.M. Sapuan, Ahmad Fudholi

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Title : Effect of sonication time on the thermal stability, moisture absorption, and biodegradation of water hyacinth (Eichhornia crassipes) nanocellulose-filled bengkuang (Pachyrhizus erosus) starch biocomposites

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Date : October 8, 2019

Reviewer #1 Comment	Author Respond and Correction
-Reviewer 1 - The presented work deals with the investigation of effect of sonication time on thermal stability, moisture absorption and biodegradation of nanocellulose water hyacinth filled bengkuang starch biocomposites produced by the solution casting method. The Authors studied the obtained materials using thermogravimetric analysis, moisture absorption, Fourier transform infrared, scanning electron microscopy and transmission electron microscopy. It was found that the biocomposite sample after vibration for 60 min revealed the highest thermal stability and exhibited low moisture absorption. The soil burial test proved that this biocomposite showed a slower biodegradation rate than the 0 min vibrated sample. It is an interesting paper of great practical importance. The topic and scope of the manuscript is suitable for the readership of this journal. I would recommend acceptance of the paper in the Journal of Materials Research and Technology after a minor revision. Some detailed comments are as following: 1). The subject matter is appropriate for the Journal of Materials Research and Technology. 2). The quality of the presentation is adequate. 3). The work contains new and original contributions. 4). Any apparent lack of clarity. 5). Any apparent errors of fact or logic. 6). Appropriate reference to previous work is given.	The revision point: 1. This is ok 2. This is ok 3. This is ok 4. This is ok 5. This is ok 6. This is ok 7. This is ok 8. This is ok 9. This is ok 10. This is ok 11. The names and surnames of the authors and its affiliation are revision. 12. We have revised and added the standard deviation in moisture absorption graphic (Figure 4). 13. We have revised and added the standard deviation in weight loss of biodegradation rate graphic (Figure 6). 14. All the spectra in Figure 5, we have added the value of (X, Y). So, we can compare the all spectra (VT 0 min, VT 15 min, VT 30 min, VT 60 min) of FTIR. For example, comparison of O-H stretching in range wavenumber 3500-3000. If we look the comparison between VT 0 min

7). The conclusions are sound and justified. 8). The abstract is informative. 9). The title reflects the contents adequately. 10). There is no material which might be omitted. 11). The names and surnames of the co-authors are not written correctly. Please provide full name for each author. Please, correct. 12). The quantitative results for the experimentally determined moisture absorption (see in Figure 4) should be given together with the standard deviation. Please, correct. 13). The quantitative results for the experimentally determined weight loss (see in Figure 6) should be given together with the standard deviation. Please, correct. 14). All the spectra shown in Figure 5 should be presented using one common X and Y axis. In the present form, it is difficult to compare the intensity and position of the peaks.	and VT 30 min, we found the point of X, Y in each spectrum. For VT 0 min (X= 3290, Y= 15) compared to the VT 60 min (X=3280, Y=23).

Reviewer #2 Comment	Author Respond and Correction
-Reviewer 2 - The paper in general is not clearly written. The paper can be published after major improvements as follows 1) It need to thoroughly checked by a native English speaker who is an expert in polymer composites for style and grammar 2) The main objective objective of this study is to determine the effect of sonoficatiob time. However, it is very confusing. It is not clear sonification of which process> This is what has been written in Introduction: "We present a novel study about the biodegradation test of 89 biocomposites. Several researchers have investigated the use of distilled 90 water to clean samples after biodegradation in soil [14–15]. However, that method is ineffective for separating soil from the sample. The soil continues to adhere to the sample due to the lowest water's energy, as substantiated by several experiments. Accordingly, this work proposes a new method for cleaning the sample. Using an ultrasonic	1. The manuscript is thoroughly revised and all possible grammatical error has been corrected via proofread professional. 2. The aim of this study to investigate the effect of vibration (sonication) time / duration during fabrication of biocomposite samples. The variation is 0, 15, 30 and 60 min. So, we want to confirmation that this study to know the properties of biocomposite affected by effect vibration during fabrication. The properties namely, thermal stability, moisture resistance and biodegradation in soil. For case of biodegradation in soil, we provide new method to remove the soil from sample after biodegradation testing. We use sonication to remove it. This is just new method. 3. The conclusion is revision. The potential application of this biocomposite is in food

bath is an effective means to separate soil from the sample after biodegradation." It seems contradicting what has been mentioned in Section 2.4 on Biocomposite fabrication 3) In Conlusion it is mentioned, The properties of the biocomposites suggest their potential application as environmentally friendly plastic for food packaging. Explain clearly which properties that the authors meant that suggest nanocellulose water hyacinth filled bengkuang starch biocomposites have potential application as environmentally friendly plastic for food packaging. 4) Why limit the sonication time to only 60 minutes ? 5) There are more corrections needed but not mention here. ALL authors must work together to improve the paper	packaging application especially in packaging bag. 4. Because, this is an optimum condition (60 min) as reported by previous report. If we use > 60 min, the biocomposite sample become broken due to sonication time. 5. All authors have a significant contribution as follow:. **Edi Syafri and Mochamad Asrofi** conducted the experiments, wrote the paper and analyzed the data. **Sudirman, Mashadi, Evi Yulianti, and Deswita** provided characterization of samples. **Hairul Abral** as the supervisor of this research provides the addition of in-depth addition analyzed the data, improved the paper quality; **R.A. Ilyas, S. M. Sapuan, and Ahmad Fudholi** as the contribution of provide standard deviation analysis and others more analysis also in revision/improvement of this manuscript.

Acknowledgement

Authors also would like to thank the Reviewers and Editors due their appropriate and constructive suggestions as well as their proposed corrections, which have been utilized in improving the quality of this manuscript.

CERTIFICATE OF EDITING

This is to certify that the paper titled Effect of sonication time on the thermal stability, moisture absorption, and biodegradation of water hyacinth (Eichhornia crassipes) nanocellulose-filled bengkuang (Pachyrhizus erosus) starch biocomposites commissioned to us by Ahmad Fudholi has been edited for English language, grammar, punctuation, and spelling by Enago, the editing brand of Crimson Interactive Inc. under Substantive Editing.

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Effect of sonication time on the thermal stability, moisture absorption, and biodegradation of ~~nanocellulose~~ water hyacinth (*Eichhornia crassipes*) nanocellulose-filled bengkuang (*Pachyrhizus erosus*) starch biocomposites

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ABSTRACT

In Indonesia, starch, ~~especially~~ particularly that obtained from bengkuang (*Pachyrhizus erosus*), is abundant and inexpensive, thereby increasing the value of bengkuang starch ~~through its role in mixing~~, which can be mixed with bioplastic-based starch. ~~Biocomposite from~~ A biocomposite comprising nanocellulose from water hyacinth (*Eichhornia crassipes*) ~~fibre~~ and bengkuang ~~(*Pachyrhizus erosus*)~~ starch was successfully fabricated using the solution casting method. ~~The nanocellulose~~ Nanocellulose content in the matrix was kept constant at 1 wt%. Moreover, ~~During~~ during fabrication, ~~a~~ the biocomposite gel was treated ~~by~~in an ultrasonic bath for 0, 15, 30, and 60 min. Further, ~~Thermogravimetric~~ thermogravimetric analysis, moisture absorption measurements, Fourier transform infrared spectroscopy, and scanning electron microscopy were performed. The

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biocomposite sample ~~with~~vibrated for 60 min ~~vibration has~~had the highest thermal stability and ~~exhibits~~exhibited low moisture absorption. The soil burial test ~~proves~~proved that this biocomposite, as opposed to 0-min vibrated samples, has a slower biodegradation rate ~~than the 0 min vibrated sample.~~. This result was supported by morphological evaluation after biodegradation, in which the 60 -min vibrated ~~sample has~~samples showed a coarse surface and low porosity formation.

Keywords: Nanocellulose, biodegradation, biocomposites, hyacinth ~~fibre~~fiber, thermogravimetric analysis.

1. INTRODUCTION

Plastic is ~~one of the materials that are~~ widely used in many applications, ~~such as~~including its use in food packaging, electronic components, and automotive dashboards. Usually, plastic is made using crude oil. However, plastic has a negative impact on the environment because it is ~~non-degradable~~nondegradable and it causes air pollution [1]. In 2010, Indonesia was responsible for the second ~~country that produces the most~~ highest level of marine plastic ~~waste in the ocean~~pollution after China. The total production of marine plastic ~~marine~~ debris globally ~~reached~~ranges 0.48–1.29 million metric tons per year [2].

One breakthrough solution to this problem ~~is~~ was the development of biodegradable plastic (bioplastic). Generally, bioplastic ~~is made from~~ comprises starch, polylactic acid, and polyvinyl alcohol. In Indonesia, starch, especially that from bengkuang~~,~~ (*Pachyrhizus erosus*), is abundant and ~~low cost~~inexpensive [3–5]. The total production of bengkuang has reached 191.5 quintals/ha per year, thereby indirectly increasing the value of bengkuang starch ~~through~~based on its ~~role in mixing~~ability to be mixed with bioplastic-based starch.

Bengkuang starch has several advantages, namely, its availability, low cost, and environmental friendliness. This starch has a high amylose content of ~~about~~approximately 30%–40% [6, 7]. However, this starch has the disadvantages of low thermal stability and high moisture absorption [7]. ~~Addition~~The addition of cellulose ~~fibre~~fiber is ~~one of the~~an alternative ~~solutions~~solution to such problems.

Cellulose ~~fibre~~fiber from water hyacinth (*Eichhornia crassipes*) is a candidate for reinforcement ~~due to~~because of its high cellulose content and abundance in ~~earth~~nature [8, 9]. Water hyacinth ~~(*Eichhornia crassipes*)~~ is a free-floating macrophyte that exhibits a fast growth rate, adaptability to a wide range of environmental conditions, and ~~a~~ high nutrient uptake capacity. Cellulose in the ~~nanometre~~nanometer range ~~is~~, called nanocellulose ~~and~~, has become prevalent in recent years. Nanocellulose has several advantages, such as being biodegradable and renewable~~,~~ and having acceptable transparency [10, 11]. Prior research established that the addition of nanocellulose in the matrix increased thermal and moisture resistance.

However, these properties depend on several factors, including porosity, the agglomeration phenomenon, and the dispersion of nanocellulose in the matrix [12]. ~~Ultrasonic treatment was~~Ultrasonication is used to reduce the formation of agglomeration and increase the dispersity of nanocellulose in ~~fibres.~~fibers. Several ~~works have~~studies reported ~~on~~ the use of ultrasonic treatment of nanocellulose ~~to reduce~~for reducing agglomeration ~~while gelatinised.~~during gelatinization. Previous researchers prepared and sonicated biocomposites from cellulose ~~fibre~~fiber-reinforced starch during ~~gelatinisation by~~gelatinization using an ultrasonic bath instrument. They claimed that sonication successfully reduced ~~fibre~~fiber agglomeration in the starch matrix [12–13]. However, their work did not reveal the biodegradation characteristic of the biocomposite sample. Note that biodegradation has an important role in biocomposites, as previously reported [14, 15].

Several researchers have investigated the use of distilled water to clean samples after biodegradation in soil [14–15]. However, ~~that~~ this method is ineffective for separating soil from ~~the sample. The soil~~samples. Soil continues to adhere to ~~the sample due to~~samples because of the lowest ~~water's~~ -energy water, as substantiated by several experiments. Accordingly, this work ~~proposes~~proposed a new method for cleaning ~~the sample.~~samples. Using an ultrasonic bath is an effective means ~~to separate~~of separating soil from ~~the sample~~samples after biodegradation. The morphological ~~characteristic~~characteristics of ~~the sample~~samples after

biodegradation ~~is~~ were observed ~~by~~via scanning electron microscopy (SEM). Thermogravimetric analysis (TGA), moisture absorption measurements, and Fourier transform infrared (FTIR) spectrometry were conducted to determine thermal stability, moisture resistance, and the functional group of the biocomposites, respectively. ~~The aim of this~~This study aimed to investigate the effect of the vibration (sonication) ~~time /~~ duration (0, 15, 30, or 60 min) during the fabrication of biocomposite samples~~. The variation is 0, 15, 30 and 60 min. So, we want to confirmation that this study to know the properties of biocomposite affected by effect vibration during fabrication. The properties namely,~~, specifically focusing on thermal stability, moisture resistance, and biodegradation in soil.

2. MATERIALS AND METHODS

2.1 Materials

Water hyacinth ~~fibre~~fiber was obtained from Payakumbuh, Indonesia. Nanocellulose was obtained from ~~the~~ water hyacinth ~~fibre was obtained by~~fiber via chemical and mechanical treatment. The lignin, hemicellulose, and cellulose contents of water hyacinth were 7%, 29%, and 43%, respectively. Bengkuang tubers were purchased from local farmers in Kuranji, Padang, Indonesia. All chemical reagents, such as distilled water, glycerol, NaOH, $NaClO_2$, CH_3COOH, and HCl, were obtained from the Metallurgy Mechanic Laboratory, Andalas University.

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Remark: Please ensure that the number of samples used for each test is clarified throughout the manuscript.

2.2. Extraction of bengkuang starch

Bengkuang tubers were peeled, cut into small pieces, and then crushed using an ice blender at 10000 rpm for 5 min to obtain the resulting porridge. The porridge was filtered using a mesh screen (200 mesh) to separate the bagasse and ~~the~~ suspension. Furthermore, ~~The~~ the suspension was precipitated for 5 h to acquire the bengkuang starch. The precipitant was dried in an oven at 50 °C for 20 h. Then, the resulting material was collected and crushed to produce dry starch powder.

2.3. Isolation of nanocellulose from water hyacinth

Water hyacinth ~~fibre~~fiber was prepared ~~following a previous report~~as previously described [9]. Dried water hyacinth ~~fibre~~fiber (15 g) was pulped ~~by~~ using 15% sodium hydroxide solution. The mixture was heated and stirred at 60 °C and 300 rpm for 4 h. Then, the ~~fibres were~~fiber was cleaned from the alkali solution with distilled water. ~~The bleaching~~ Bleaching ~~process~~ was performed ~~with~~ using sodium chlorite and acetic acid (4:1). This process was conducted at 60 °C and 300 rpm for 2 h. ~~It~~The mixture was rinsed ~~by~~with distilled water to obtain the cellulose suspension. Next, 5 M HCl was added to the suspension for acid hydrolysis at 60 °C and 300 rpm for 20 h. ~~HCL~~HCl was used to isolate nanocellulose from the cellulose ~~fibre.~~fiber. Then, the products were ~~neutralised~~neutralized using distilled water before ultrasonication. An ultrasonic crusher was ~~utilised~~utilized to produce nanocellulose from water hyacinth. This process was conducted at 600 W for 1 h.

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Tip: Redundancy: Redundant phrases make a sentence wordy. Being economical in writing enhances the clarity and readability of the sentence. Here, the use of both "by" and "using" is redundant.

2.4. Biocomposite fabrication

A total of 10 g of bengkuang starch was mixed with 100 ml of distilled water in a glass beaker. This mixture was ~~homogenised by~~homogenized using an ultrasonic ~~homogeniser~~homogenizer at 8000 rpm for 5 min. During ~~homogenisation~~homogenization, 1% NWHF and 2 ml of glycerol were ~~added~~ slowly added to the mixing solution. A constant fraction of nanocellulose (1 wt%) was used as reinforcement in the bengkuang starch matrix ~~.~~ Then, the mixing solution was heated and stirred at 60 °C and 500 rpm for 30 min (until ~~gelatinised~~gelatinized). The biocomposite gel (70 gr) was poured in a ~~petri~~Petri dish (~~d=~~15 cm diameter). The gel was ~~treated by~~incubated in an ultrasonic bath for 0, 15, 30 ~~and~~ , or 60 min and labeled as ~~labelled samples for~~ VT 0 (untreated), VT 15, VT 30, and VT 60, respectively. Subsequently, the biocomposite was dried in a drying oven at 50 °C for 20 h.

2.5. ~~Characterisation~~Characterization

2.5.1. ~~Scanning Electron Microscopy~~SEM

The morphology of the untreated and treated water hyacinth ~~fibres~~fibers as well as the morphological characteristic of all biocomposites tested after

biodegradation in soil were observed ~~by~~ using ~~SEM~~a Hitachi 3400 N~~.~~ scanning electron microscope. The operation voltage was 10 kV. The fracture surface of the biocomposite was studied through ~~the SEM~~a VEGA3 TESCAN SEM instrument at room temperature and 10 kV. ~~-~~All samples were coated with gold ~~by~~ using an argon plasma ~~metalliser~~metalizer (sputter coater K575X~~) (~~, Edwards Limited, Crawley, ~~United Kingdom~~UK) to avoid charging.

2.5.2. Transmission Electron Microscopy

~~Transmission electron microscopy~~ **(TEM)**

A JEM-JEOL 1400 TEM instrument was operated at 100 keV to observe the nanocellulose ~~water hyacinth fibre.~~fibers. The nanocellulose suspension was cast in a carbon-coated grid and then directly observed at room temperature.

2.5.3. Thermogravimetric Analysis

All samples were tested ~~by~~using a TGA/DTG 60 system (serial no. C30565000570) to determine the thermal degradation point at each stage. The test was conducted ~~in~~over the temperature range of 30 °C–550 °C under a nitrogen atmosphere. The heating rate was maintained at 10 °C/min. The weight of all tested samples was 5–7 mg.

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Remark: Abbreviations need to be spelled out at their first usage in the abstract and the main text and used consistently thereafter. Please check if you need to expand all the abbreviations in the text at their first usage. If they are mentioned in the journal's list of accepted abbreviations, they do not need to be spelled out.

2.5.4. Moisture Absorption

All tested samples were cut into 1 cm × 3 cm pieces and dried in a drying oven to a constant weight. The initial (W_o) and final weights ~~are (W_o) and~~ (W_t)~~, respectively.~~) were recorded. Moisture absorption was ~~performed~~examined in a moisture chamber (relative humidity [RH]: 75%) at 25 °C. The ~~final weight (Wt) is the final weighing of the sample every 30 min. The percentage of~~percent moisture absorption was calculated ~~by~~ using ~~a previous~~the following equation [12~~].~~]:

$$Moisture\ Absorption = \frac{W_t - W_o}{W_o} \times 100\%. \quad (1)$$

2.5.5. ~~Fourier Transform Infrared~~ FTIR ~~characterisation~~spectroscopy

FTIR spectroscopy was used to determine the functional groups of all samples. The FTIR spectrum of all tested samples was recorded ~~by~~ using a Perkin-Elmer Frontier FTIR instrument with a resolution of 4 cm~~-~~$^{-1}$. The scanned wavenumber range was 600–4000 cm~~-~~$^{-1}$.

2.5.6. Soil Burial Test

The biodegradation test was conducted by burying the ~~sample~~ samples in soil that was purchased from PT. Mahesa Mutiara Tani (Kompos Nesia~~),~~, Bogor, Indonesia~~.~~). The soil contained organic carbon (34.25%), ~~carbon-to-nitrogen ratio (17.15%),~~ nitrogen (2%), ferrous (7.9%), pH (7.33), diphosphorus ~~pentaoxide~~pentoxide (0.62%), other materials (e.g., plastic, glass, gravel~~, etc.~~ = 0.1~~%)~~%), and several colonies in 1 gram of soil (8 × 10^{14} CFU~~).~~), and its carbon-to-nitrogen ratio was 17.15%.

Control sample: The sample was cut into 10 mm × 10 mm pieces ~~for each variation~~ and then dried in a drying oven at 50~~-~~°C for 20 h to a constant weight. Then, the sample was subjected to ~~an~~ ultrasonic bath treatment for 5 min and dried in an oven at 50~~-~~°C for 20 h. Subsequently, the sample was weighed ~~by~~ using a precision balance (accuracy = 0.001). The weight loss of the control sample was ~~obtained from~~calculated as the difference between ~~the initial sample weight~~W_o before ~~the~~ ultrasonic bath treatment and ~~the final weight~~W_t after the ~~said bath~~treatment.

~~Burial sample~~Buried samples: All samples for the burial test measured 10 mm × 10 mm ~~for each variation.~~. The samples were dried in a drying oven at 50~~-~~°C for 20 h to a constant weight and weighed ~~with~~using a precision balance to obtain ~~the initial weight~~W_o before burial in soil. Then, the test was conducted in a square container (10 cm × 10 cm) ~~that contains~~containing soil ~~with relative humidity~~an RH and temperature of 60%–65% and 25 °C, respectively. ~~All tested samples were non-vibrated (0 min), vibrated at 15 min, vibrated at 30 min and vibrated at 60 min. The~~After vibration, biodegradation was observed ~~for~~after 3, 7, and 15 days of burial in soil.

After ~~the~~ burial, all samples were collected and cleaned ~~by~~ using a paintbrush. Then, the samples ~~underwent~~were subjected to ultrasonic bath treatment for 5 min to remove the soil that adhered to the ~~biocomposite sample.~~biocomposites. The samples were dried in a drying oven at 50 ~~–~~ °C for 20 h to a constant weight. ~~Afterwards~~Afterward, they were weighed ~~with~~using an analytical balance. The ~~percentage~~percent weight loss of the sample due to soil burial was calculated according to the different weights before and after the burial test. ~~Percentage~~The percent weight loss due to soil was calculated ~~by~~using the following equation ~~below~~:

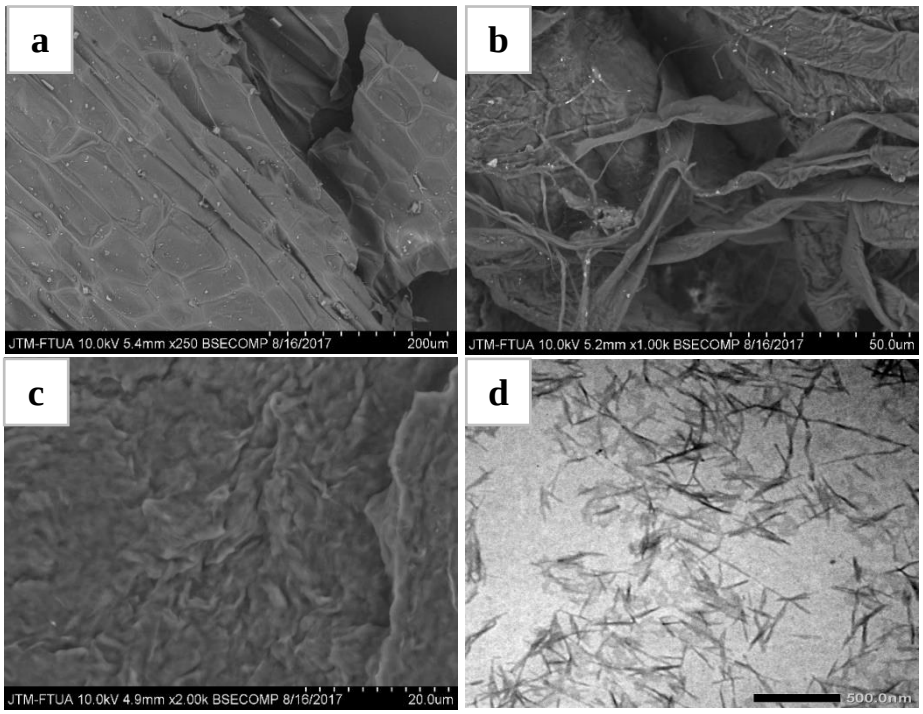
$$W_{degradation} = W_{burial\ test} - W_{control}. \quad (2)$$

3. RESULTS AND ~~DISCUSSIONS~~ DISCUSSION

3.1 Morphological characteristics of untreated and treated ~~fibre~~fibers

Figure 1 shows the morphological features of water hyacinth ~~fibre~~fiber before and after treatment. Figure 1a displays the raw water hyacinth ~~fibre~~fiber (untreated), which ~~has~~had a smooth surface, and some microfibrils ~~are~~were still bound. Untreated ~~fibre~~fiber contains lignin substances, ~~-~~ such as waxes and oils [16, 17]. Bleaching successfully ~~destroys~~destroyed several microfibril ~~bondings~~bonds, as presented in Figure 1b, because of the broken linkage between lignin and hemicellulose. A similar phenomenon was reported by a previous study [9, 18]. In the acid hydrolysis process (Figure 1c), the ~~fibre~~fiber was ~~depolymerised and became~~depolymerized into short individual microfibrils with a diameter and length of 2 and 6 microns, respectively. ~~The nanocellulose~~Nanocellulose was obtained ~~from~~via ultrasonication for 1 h. Figure 1d displays the distribution of cellulose ~~fibres~~fibers in the ~~nanometre~~nanometer range. Cellulose ~~fibres~~fibers were dispersed homogeneously, as shown in the TEM images (Figure 1d), ~~and measured~~measuring 15 nm in diameter and 147 nm in length. This outcome is ~~due~~attributable to the high intensity of the power ultrasound, resulting in short cellulose ~~fibres~~fibers in the ~~nanometre area~~nanometer range [19, 20].

Figure 1. Water hyacinth ~~fibre~~fiber before and after treatment ~~using~~



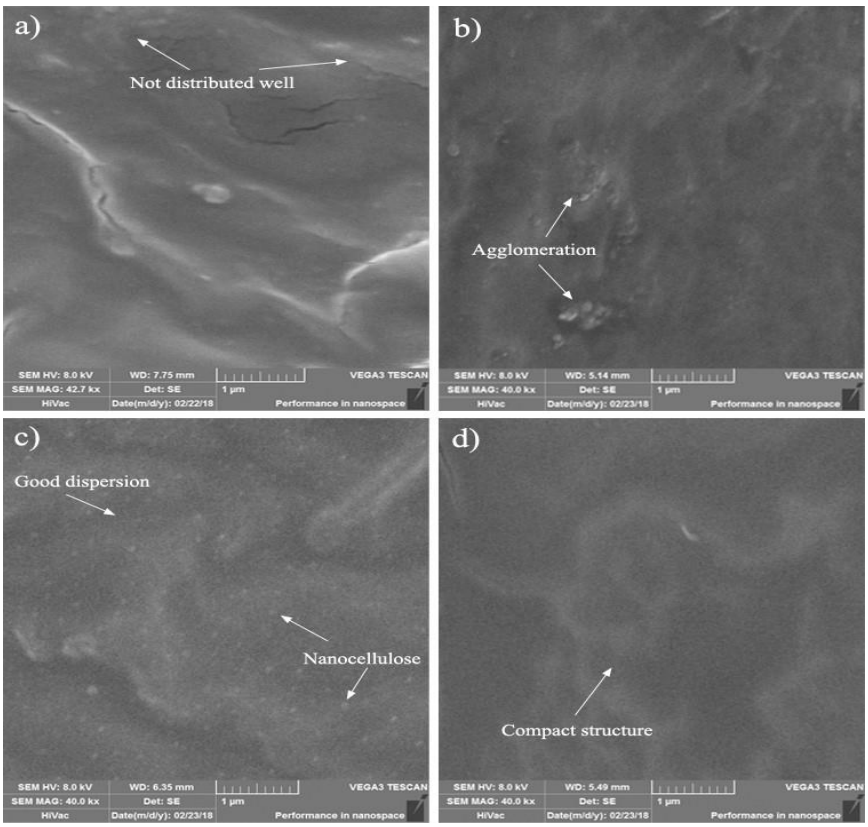
~~FESEM:.~~ (a) ~~raw fibre~~Raw fiber (untreated), (b) bleached ~~fibre~~fiber, (c) fiber subjected to acid hydrolysis ~~fibre~~; and ~~using TEM:~~ (d) ~~ultrasonicated fibre~~fiber subjected to ultrasonication.

3.2. Fracture Surface of the Biocomposites

Figure 2 shows the fracture surface of untreated and treated biocomposite ~~film.~~films. Figure 2a displays the fracture morphology of the VT 0 min sample (untreated). The cellulose ~~fibre~~fibers in bengkuang starch ~~was~~were not distributed well. Thus, the mixing of starch and cellulose was not homogeneous during fabrication [21]. This ~~case affected~~explained the low thermal stability of the untreated biocomposite. ~~Poor~~A poor distribution of nanocellulose ~~would affect~~affects thermal stability in terms of interfacial adhesion between the nanocellulose and starch ~~biopolymer.~~biopolymers. That is, the strong interfacial adhesion between nanocellulose and starch corresponded to the high thermal stability of the biocomposites because a

high temperature is required to break the ~~bond~~bonds between nanocellulose and starch ~~biopolymer~~biopolymers.

The VT 15 min sample also ~~shows~~exhibited no significant ~~effect on~~ fracture morphology. The agglomeration phenomenon ~~still appears~~remains apparent in this sample (Figure 2b). According to a previous report, agglomeration and poor dispersion of cellulose ~~fibres~~fibers lead to decreased thermal and moisture resistance [22]. The different fracture morphologies are shown in Figures 2c and 2d for the VT 30 min and VT 60 min samples, respectively. In ~~this case~~these cases, the samples ~~display~~displayed good dispersion and compact ~~structure. This fact is~~structures. These findings are probably ~~due~~attributable to the kinetic energy from the ultrasonic bath, leading to improved interfacial bonding between the ~~fibre~~fibers and matrix [12, 13]. This treatment also ~~results~~resulted in good dispersion of the nanocellulose in the matrix and ~~reduces~~reduced the free OH bonding between the ~~fibre~~fibers and ~~the~~ matrix. This phenomenon was similar to that observed by Asrofi et al. [6] ~~on~~concerning the effect of ultrasonic vibration during processing on the



mechanical properties of a water hyacinth ~~nanofibre~~nanofiber cellulose-reinforced thermoplastic starch bionanocomposite.

Figure 2. Fracture surface of all biocomposite samples~~:~~. Samples were vibrated for (a) ~~VT~~ 0 ~~min~~, (b) ~~VT~~ 15 ~~min~~, (c) ~~VT~~ 30 ~~min and~~, or (d) ~~VT~~ 60 min.

3.3. Thermal Stability

The thermal characteristics of the biocomposite samples with various ~~VT~~vibration times were presented ~~on the TG~~using TGA (Figure 3a) and DTG (Figure 3b) ~~curve.~~curves. Two main degradation areas were found in accordance with the results from previous research [23–25]. The first region of degradation ~~was~~reflected the initial degradation of all biocomposite samples ~~below~~at temperatures of less than 100 °C as indicated by the ~~TG~~TGA and DTG curves. This result ~~indicated an~~revealed increasing weight loss ~~percentage~~ compared with ~~the initial weight~~W_0 before testing. The percent weight loss values of the biocomposite in the first region ~~was~~were 11% (VT 0 min), 9% (VT 15 min), 9% (VT 30 min), and 7% (VT 60 min). This phenomenon was ~~due~~attributable to the moisture content in all samples [8, 25]. These results were supported by the DTG curve (Figure 1b), which ~~shows~~reveals the presence of small peaks in the temperature range of 50 °C–100 °C.

The second region (250 °C–350 °C) involved ~~a major percentage~~significant weight loss ~~where all samples have a weight loss above~~exceeding 50~~%.~~% for all samples. In this condition, the ~~structure~~structures of starch, glycerol, and nanocellulose ~~fibres broke down~~fibers were degraded [12, 26]. The DTG curve (Figure 1b) ~~shows~~exhibits a sharp peak due to the significant weight reduction. The degradation ~~temperature~~temperatures for all samples in this area was 305 °C, 309 °C, 311 °C, and 318 °C for VT 0, VT 15, VT 30, and VT 60 min, respectively. ~~Biocomposite~~Untreated biocomposite samples ~~with~~ (VT 0) min ~~have~~had lower degradation temperatures. This outcome indicated fiber agglomerated ~~fibre~~ and porosity formation in the matrix that resulted in

reduced thermal stability [4]. Thermal stability and moisture resistance increased with ~~longer~~prolonged ultrasonic vibration ~~time~~.

Conversely, ~~VT improves~~increased vibration time improved the thermal stability of the biocomposite samples. This result was supported by the increase in the degradation temperature of the treated ~~sample~~samples compared with that of the untreated sample due to the improved interfacial adhesion between the ~~fibre~~fibers and ~~the~~ matrix as a result of the kinetic energy of the ultrasonic bath. Good adhesion ~~bonding~~ was indicated by the scarcity of free OH between the ~~fibre~~fibers and ~~the~~ matrix [6, 8, 12].

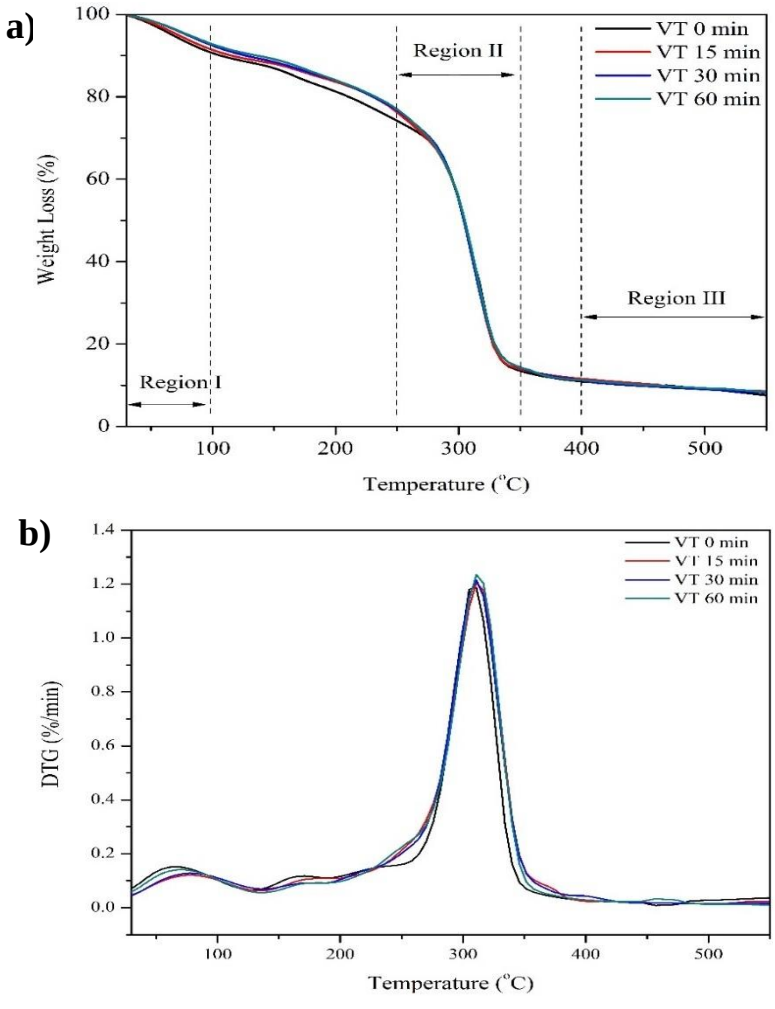


Figure 3. (a) ~~TG~~Thermogravimetric analysis and (b) DTG ~~curve~~curves of all biocomposite ~~sample~~samples

In the third region (> ~~-~~400 ~~-~~°C), all samples were completely decomposed ~~and became~~, becoming ash [25, 26]. However, the treated samples ~~have~~had higher thermal stability than untreated samples because of the kinetic energy of the ultrasonic bath, which ~~was able~~allowed the fibers to disperse ~~the fibres~~ evenly in the matrix. Other ~~impacts~~effects of the bath ~~include~~included good dispersion of the ~~fibre~~fibers in the matrix, a compact structure, and improved interfacial bonding between the ~~fibre~~fibers and ~~the~~ matrix. These thermal results are supported by SEM data and are similar to those reported in previous studies [12–13].

3.4. Moisture Absorption

Figure 4 shows the effect of ~~VT~~the vibration time on the percent moisture absorption ~~percentage~~ of the biocomposite samples. The test was conducted for 8 h under conditions of RH ~~of~~ = 75% and ~~temperature of~~ 25 °C. All biocomposite samples ~~began to be~~were saturated ~~under~~in less than 5 h. The ~~biocomposite sample VT 0 min has a~~percent moisture absorption of the VT 0 min biocomposite sample was 22%. This phenomenon was ~~due~~attributable to the ~~non-homogeneous~~nonhomogeneous dispersion of nanocellulose in the starch matrix. Another reason was the hydrophilic nature of starch and cellulose ~~fibre~~fibers [13].

These results differed from those ~~with~~of treated biocomposite samples~~. The treated biocomposites have lower~~, which exhibited less moisture absorption. For example, in the VT 60 min ~~sample~~samples, the percent moisture absorption ~~percentage decreased by~~ was 4.5% ~~relative to~~lower than that of the VT 0 min ~~counterpart.~~samples. This outcome occurred because the kinetic energy produced by the ultrasonic bath ~~was able to break down the~~could reverse fiber agglomeration ~~fibres~~, homogenously spread cellulose ~~fibres~~fibers in the starch matrix, and create a strong hydrogen bonding interaction between nanocellulose and the

starch biopolymer. Accordingly, it was difficult for water molecules ~~were difficult~~ to diffuse into the matrix because of the formation of the tortuous path made by nanocellulose, ~~whose~~the high moisture ~~contents~~content of which created a barrier effect on the movement of water molecules through the biocomposite [12–13]. This result was supported by the FTIR ~~characterisation~~characterization at a ~~wave number~~wavenumber of 1600 cm^{-1}, ~~indicates OH group~~indicated water absorption. ~~–~~by OH groups. Given the chemical similarity between nanocellulose and the starch biopolymers, the interfacial defects were reduced, thereby generating firm resistance to the flow of water molecules.

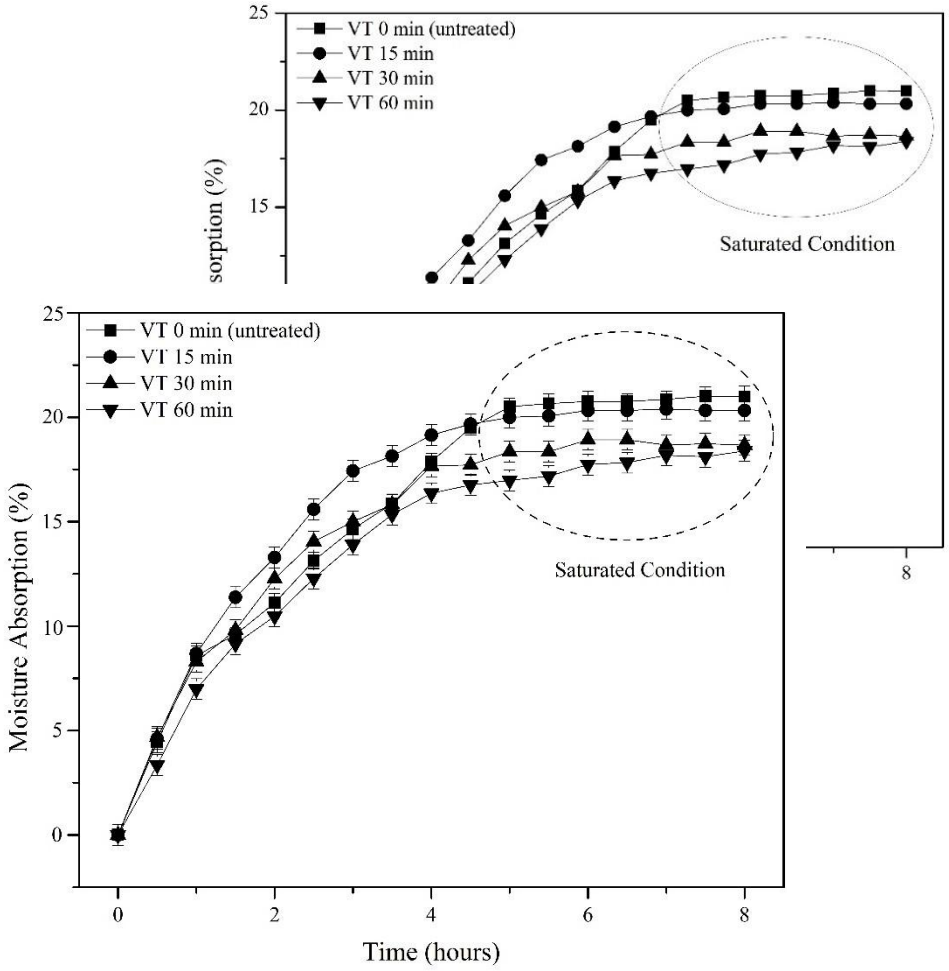


Figure 4. Moisture absorption of all tested biocomposite samples. Samples were subjected to ultrasonication for 0, 15, 30, or 60 min. VT: vibration time.

3.5. Functional group analysis

Figure 5 shows the FTIR spectrumspectra of untreated and treated biocomposites. Three main peaks occurred at different wavenumbers (Figure 5). The peak was in the wavenumber of), namely 3000 cm^{-1} (stretch (O-H stretching), 2900 cm^{-1} (stretch (C-H) stretching), and 1600 cm^{-1} (OH water absorption groupby OH groups) [27, 28].

The O-H stretching group appearsappeared at 3000 cm^{-1}, and the biocomposite showsdisplayed a shift in to higher wavenumbers in a higher direction.. This result indicated an interaction between the matrix and fibrefibers that generated a new hydrogen bonding formationbonds [28]. The transmittance values of the untreated and treated biocomposites varied. Long VT corresponded to a vibration times resulted in high transmittance valuevalues (Figure 5) because of the greater OH stretch. Thestretching. Good hydrogen bonding was noted between the matrix and fibre forms a good hydrogen bondfibers (few occurrences of free OH bonding) [12], thereby producing satisfactory tensile strength due to the good structure formation of OH molecules (crystalline) [6, 12].

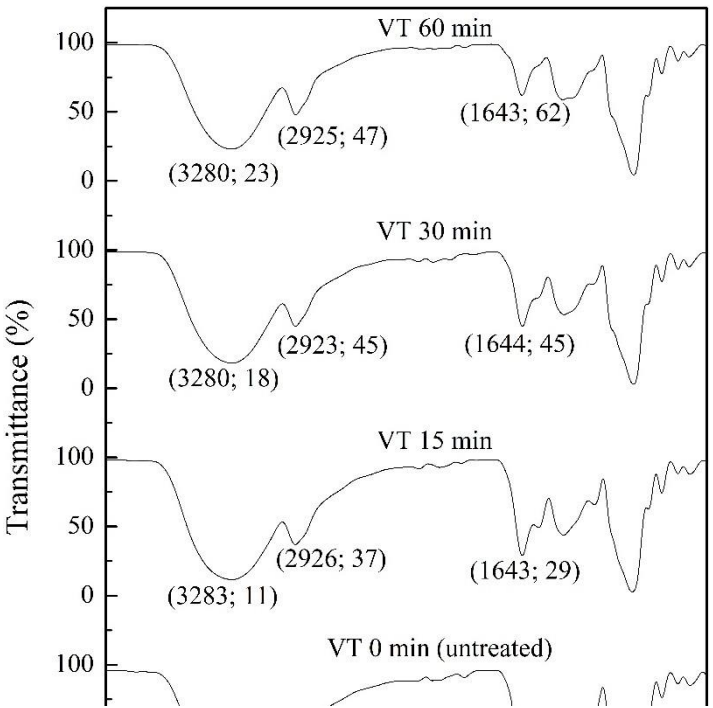


Figure 5. Functional ~~group~~groups of all tested biocomposite samples. VT: vibration time.

In addition, ~~the~~ C-H stretching ~~appears~~appeared at ~~the wave number of~~ approximately 2900 cm^{-1} in all biocomposite samples. Thus, all samples ~~contain~~contained saturated aliphatic components, as reported by previous researchers [29]. Another phenomenon also ~~appears~~appeared around ~~the wavenumber of~~ 1600 cm^{-1}. In this area, OH water absorption appears in each biocomposite sample. The transmittance value increases with the duration of ~~VT.~~vibration. The transmittance values of ~~biocomposite samples of~~the VT 0, VT 15, VT 30, and VT 60 min biocomposite samples were 25%, 29%, 45%, and 62%, respectively. These results suggest that untreated biocomposite samples ~~have higher~~exhibit greater water absorption than treated samples [4, 28]. This outcome was supported by the moisture absorption test~~, where~~ results, in which the VT 0 min ~~sample~~ samples had the highest ~~percentage of~~percent moisture absorption. However, treated biocomposite samples ~~have~~displayed lower moisture absorption. ~~Because the~~The kinetic energy of the ultrasonic bath ~~was able to break down~~could degrade the agglomerated nanocellulose ~~fibres~~fiber and spread them evenly in the matrix [12]. This phenomenon makes it difficult ~~to diffuse~~ for water molecules ~~in~~to diffuse into the matrix. A similar outcome was reported by previous research [6, 12].

Commented [6]: Page: 16
Tip: Minus sign: A minus sign is preferred over hyphen to indicate negative values.

3.6. Soil Biodegradation

Investigating soil biodegradation ~~behaviour~~behavior is crucial for ~~the utilisation of~~utilizing biocomposites in the environment. Soil biodegradation involves the degradation of materials by the action of microorganisms,

fungi, bacteria, or other ~~biological means~~organisms that live in soil. Figure 6 shows the effect of ~~VT~~the vibration time on the biodegradation rates of all biocomposite samples. Compared with treated samples, ~~Untreated~~ untreated biocomposites ~~have a higher percentage of~~exhibited greater weight loss ~~compared with treated samples~~. The percent weight loss on ~~the 3rd~~ day 3 ~~of the~~for untreated ~~sample~~samples was 9.55~~%. This reduction also occurred on the 7th~~%, and ~~15th~~ the values on days~~. The weight loss on the 7th~~ 7 and ~~15th days~~15 were 30.06% and 31.21%, respectively. This outcome was supported by ~~Figures~~the results presented in Figures 7a and 7b, ~~where~~in which the morphological structure depicts a large hole with an irregular configuration. This phenomenon may be ~~due~~attributable to ~~fibre~~fiber clumping in the matrix [8].

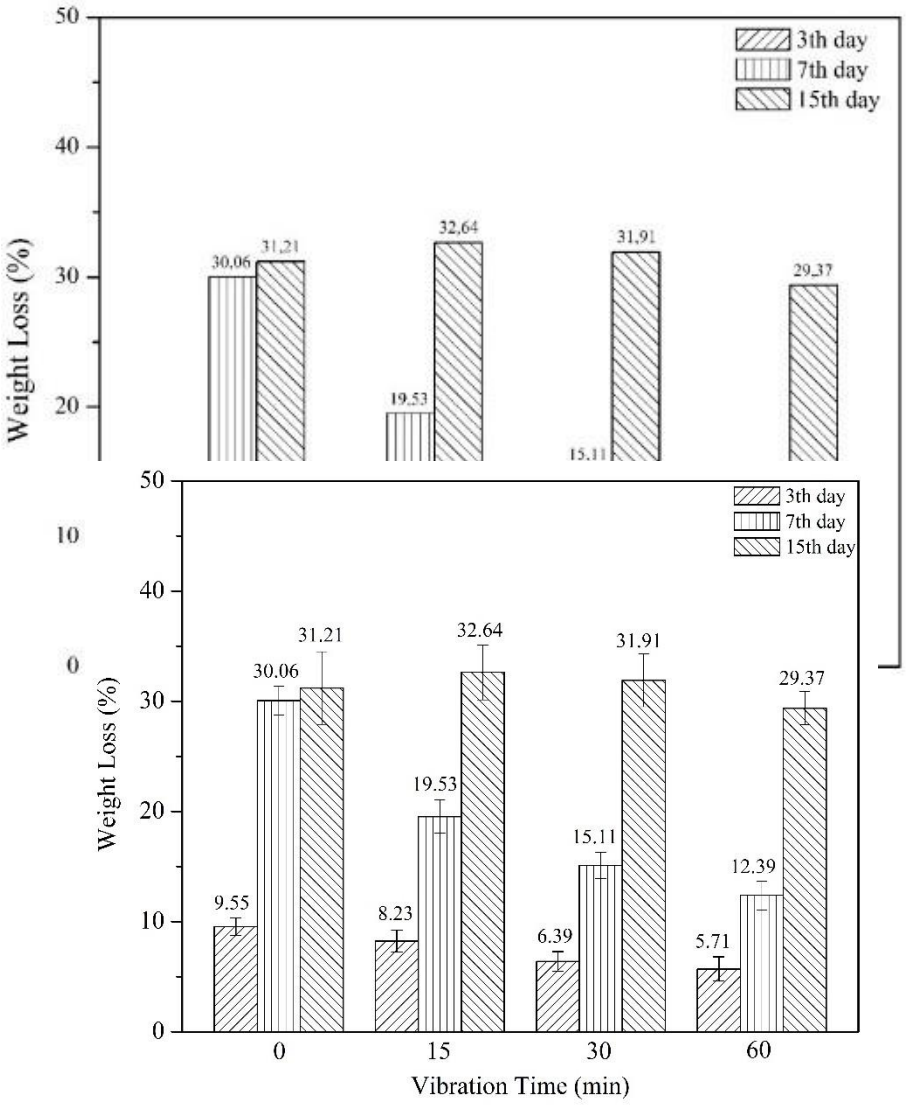


Figure 6. Biodegradation rates of all tested biocomposite samples

~~This value was different with~~ These values differed from those of VT 60 min samples. In ~~this sample,~~these samples, the percent weight loss ~~was~~values were 5.71% ~~(3rd~~ (day 3), 12.39% ~~(7th~~ (day~~)~~ 7), and 29.37% (~~15th~~ day 15). Thus, VT 60 min ~~has~~samples had a slower degradation rate than the untreated ~~sample.~~samples. This phenomenon was ~~due~~attributable to the ~~the~~ kinetic energy from the ultrasonic bath ~~that~~, which distributed the ~~fibres~~fibers homogeneously within the matrix [6]. The good fiber distribution ~~of fibre~~ in the matrix affected moisture, microorganisms, and other elements~~,~~ such that ~~diffusing them~~their diffusion into the matrix was difficult, as reported by previous research [14, 15, 30–32]. These results ~~are~~were supported by the SEM ~~imaging (Figures~~ findings (Figure 7c and 7d), ~~where~~in which the biocomposite structure was compact and less porous. The ~~effect of~~ vibration time also decreased the biodegradation rate. These phenomena ~~can~~may be correlated with the water absorption properties of the biocomposite film, ~~where the~~in which untreated film tends to absorb more water than ~~the~~ treated film, thereby making the former more prone to ~~microorganism~~ attacks~~. Microorganisms~~ by microorganisms, which can attack the starch biopolymer in the presence of a water medium.

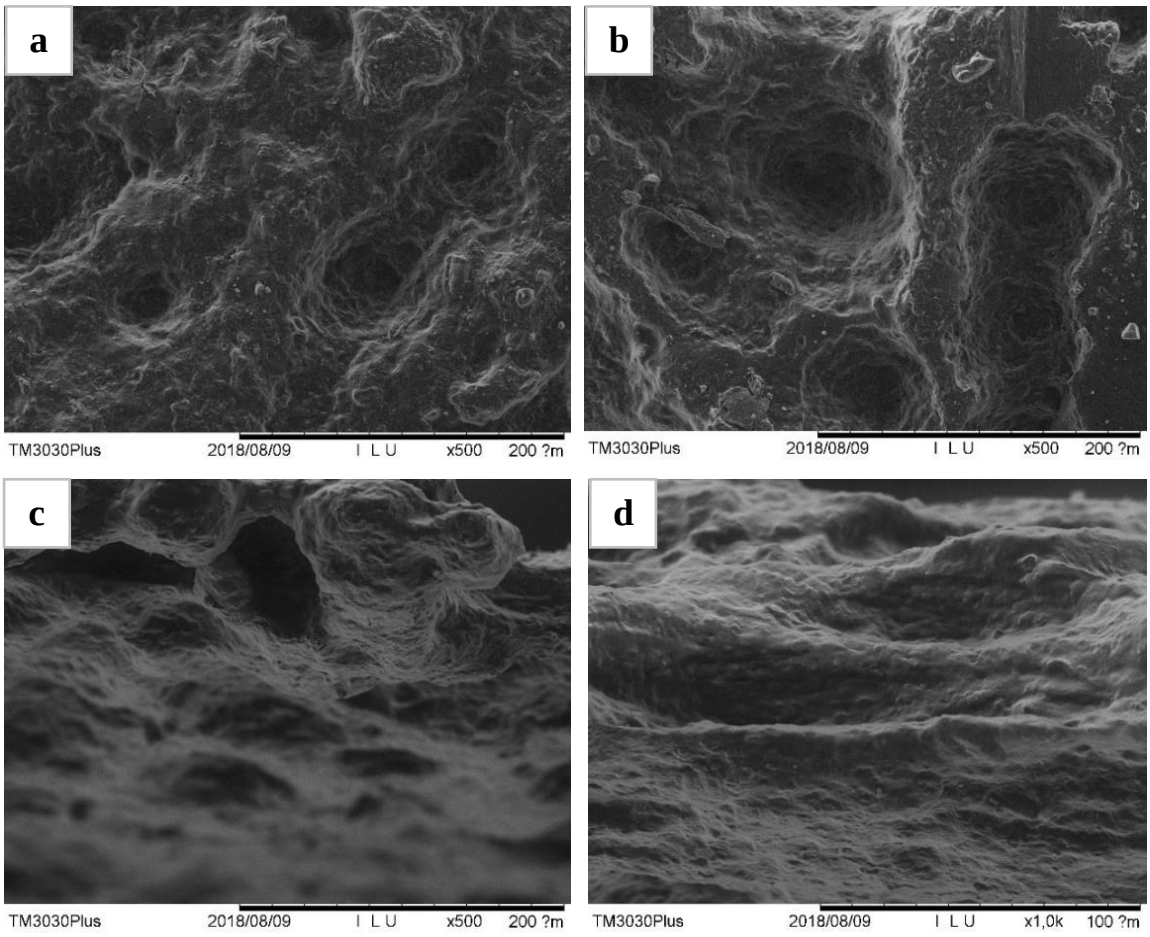


Figure 7. Morphological characteristics of biocomposite samples after biodegradation in soil~~:~~. (a) ~~VT 0 min (3rd~~ Untreated (day 3), (b) ~~VT 0 min (15th~~ untreated (day 15), (c) ~~VT~~vibrated for 60 min (~~3rd~~ day~~)~~ 3), and (d) ~~VT~~vibrated for 60 min (~~15th~~ day~~)~~ 15).

4. CONCLUSION

Thermal stability and moisture resistance were successfully improved by vibration treatment ~~from~~using an ultrasonic bath instrument. Kinetic energy from the ultrasonic bath reduced the free OH bonding between the ~~fibre~~fiber and matrix. The ~~maximum condition was achieved in the~~best results were obtained for VT 60 min biocomposite ~~sample. An increase in~~samples. Increasing the vibration time also ~~inhibited~~reduced the degradation rate of biocomposites in the soil. The soil burial test revealed that ~~these~~ the vibrated biocomposites ~~have~~had slower biodegradation rates ~~compared with~~than the ~~0 min vibrated sample.~~samples. The properties of

the biocomposites suggest their potential application as environmentally friendly ~~plastic~~plastics for food packaging. The potential application of this biocomposite is ~~in~~ food packaging ~~application~~, especially ~~in~~as packaging ~~bag~~bags.

ACKNOWLEDGMENT

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CONFLICTS OF INTEREST

The authors declare no conflict of interest regarding the publication of this manuscript. This manuscript has not been published and is not under ~~considered~~ consideration for publication elsewhere. The authors certify that neither the manuscript nor its main contents have ~~already~~ been published or submitted for publication in another journal.

Effect of sonication time on the thermal stability, moisture absorption, and biodegradation of water hyacinth (*Eichhornia crassipes*) nanocellulose-filled bengkuang (*Pachyrhizus erosus*) starch biocomposites

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ABSTRACT

In Indonesia, starch, particularly that obtained from bengkuang (*Pachyrhizus erosus*), is abundant and inexpensive, thereby increasing the value of bengkuang starch, which can be mixed with bioplastic-based starch. A biocomposite comprising nanocellulose from water hyacinth (*Eichhornia crassipes*) and bengkuang starch was successfully fabricated using the solution casting method. Nanocellulose content in the matrix was kept constant at 1 wt%. Moreover, during fabrication, the biocomposite gel was treated in an ultrasonic bath for 0, 15, 30, and 60 min. Further, thermogravimetric analysis, moisture absorption measurements, Fourier transform infrared spectroscopy, and scanning electron microscopy were performed. The biocomposite sample vibrated for 60 min had the highest thermal stability and exhibited low moisture absorption. The soil burial test

proved that this biocomposite, as opposed to 0-min vibrated samples, has a slower biodegradation rate. This result was supported by morphological evaluation after biodegradation, in which the 60-min vibrated samples showed a coarse surface and low porosity formation.
Keywords: Nanocellulose, biodegradation, biocomposites, hyacinth fiber, thermogravimetric analysis

1. INTRODUCTION

Plastic is widely used in many applications, including its use in food packaging, electronic components, and automotive dashboards. Usually, plastic is made using crude oil. However, plastic has a negative impact on the environment because it is nondegradable and it causes air pollution [1]. In 2010, Indonesia was responsible for the second highest level of marine plastic pollution after China. The total production of marine plastic debris globally ranges 0.48–1.29 million metric tons per year [2].

One breakthrough solution to this problem was the development of biodegradable plastic (bioplastic). Generally, bioplastic comprises starch, polylactic acid, and polyvinyl alcohol. In Indonesia, starch, especially that from bengkuang (*Pachyrhizus erosus*), is abundant and inexpensive [3–5]. The total production of bengkuang has reached 191.5 quintals/ha per year, thereby indirectly increasing the value of bengkuang starch based on its ability to be mixed with bioplastic-based starch. Bengkuang starch has several advantages, namely its availability, low cost, and environmental friendliness. This starch has a high amylose content of approximately 30%–40% [6, 7]. However, this starch has the disadvantages of low thermal stability and high moisture absorption [7]. The addition of cellulose fiber is an alternative solution to such problems.

Cellulose fiber from water hyacinth (*Eichhornia crassipes*) is a candidate for reinforcement because of its high cellulose content and abundance in nature [8, 9]. Water hyacinth is a free-floating macrophyte that exhibits a fast growth rate, adaptability to a wide range of environmental conditions, and high nutrient uptake capacity. Cellulose in the nanometer range, called nanocellulose, has become prevalent in recent years. Nanocellulose has several advantages, such as being biodegradable

and renewable and having acceptable transparency [10, 11]. Prior research established that the addition of nanocellulose in the matrix increased thermal and moisture resistance.

However, these properties depend on several factors, including porosity, the agglomeration phenomenon, and the dispersion of nanocellulose in the matrix [12]. Ultrasonication is used to reduce the formation of agglomeration and increase the dispersity of nanocellulose in fibers. Several studies reported the use of ultrasonic treatment of nanocellulose for reducing agglomeration during gelatinization. Previous researchers prepared and sonicated biocomposites from cellulose fiber-reinforced starch during gelatinization using an ultrasonic bath instrument. They claimed that sonication successfully reduced fiber agglomeration in the starch matrix [12–13]. However, their work did not reveal the biodegradation characteristic of the biocomposite sample. Note that biodegradation has an important role in biocomposites, as previously reported [14, 15].

Several researchers have investigated the use of distilled water to clean samples after biodegradation in soil [14–15]. However, this method is ineffective for separating soil from samples. Soil continues to adhere to samples because of the lowest-energy water, as substantiated by several experiments. Accordingly, this work proposed a new method for cleaning samples. Using an ultrasonic bath is an effective means of separating soil from samples after biodegradation. The morphological characteristics of samples after biodegradation were observed via scanning electron microscopy (SEM). Thermogravimetric analysis (TGA), moisture absorption measurements, and Fourier transform infrared (FTIR) spectrometry were conducted to determine thermal stability, moisture resistance, and the functional group of the biocomposites, respectively. This study aimed to investigate the effect of the vibration (sonication) duration (0, 15, 30, or 60 min) during the fabrication of biocomposite samples, specifically focusing on thermal stability, moisture resistance, and biodegradation in soil.

2. MATERIALS AND METHODS

2.1 Materials

Water hyacinth fiber was obtained from Payakumbuh, Indonesia. Nanocellulose was obtained from water hyacinth fiber via chemical and mechanical treatment. The lignin, hemicellulose, and cellulose contents of water hyacinth were 7%, 29%, and 43%, respectively. Bengkuang tubers were purchased from local farmers in Kuranji, Padang, Indonesia. All chemical reagents, such as distilled water, glycerol, NaOH, $NaClO_2$, CH_3COOH, and HCl, were obtained from the Metallurgy Mechanic Laboratory, Andalas University.

2.2. Extraction of bengkuang starch

Bengkuang tubers were peeled, cut into small pieces, and then crushed using an ice blender at 10000 rpm for 5 min to obtain the resulting porridge. The porridge was filtered using a mesh screen (200 mesh) to separate the bagasse and suspension. Furthermore, the suspension was precipitated for 5 h to acquire the bengkuang starch. The precipitant was dried in an oven at 50°C for 20 h. Then, the resulting material was collected and crushed to produce dry starch powder.

2.3. Isolation of nanocellulose from water hyacinth

Water hyacinth fiber was prepared as previously described [9]. Dried water hyacinth fiber (15 g) was pulped using 15% sodium hydroxide solution. The mixture was heated and stirred at 60°C and 300 rpm for 4 h. Then, the fiber was cleaned from the alkali solution with distilled water. Bleaching was performed using sodium chlorite and acetic acid (4:1). This process was conducted at 60°C and 300 rpm for 2 h. The mixture was rinsed with distilled water to obtain the cellulose suspension. Next, 5 M HCl was added to the suspension for acid hydrolysis at 60°C and 300 rpm for 20 h. HCl was used to isolate nanocellulose from the cellulose fiber. Then, the products were neutralized using distilled water before ultrasonication. An ultrasonic crusher was utilized to produce nanocellulose from water hyacinth. This process was conducted at 600 W for 1 h.

2.4. Biocomposite fabrication

A total of 10 g of bengkuang starch was mixed with 100 ml of distilled water in a glass beaker. This mixture was homogenized using an ultrasonic homogenizer at 8000 rpm for 5 min. During homogenization, 1% NWHF and 2 ml of glycerol were slowly added to the mixing solution. A constant fraction of nanocellulose (1 wt%) was used as reinforcement in the bengkuang starch matrix. Then, the mixing solution was heated and stirred at 60°C and 500 rpm for 30 min (until gelatinized). The biocomposite gel (70 gr) was poured in a Petri dish (15 cm diameter). The gel was incubated in an ultrasonic bath for 0, 15, 30, or 60 min and labeled as VT 0 (untreated), VT 15, VT 30, and VT 60, respectively. Subsequently, the biocomposite was dried in a drying oven at 50°C for 20 h.

2.5. Characterization

2.5.1. SEM

The morphology of the untreated and treated water hyacinth fibers as well as the morphological characteristic of all biocomposites tested after biodegradation in soil were observed using a Hitachi 3400 N scanning electron microscope. The operation voltage was 10 kV. The fracture surface of the biocomposite was studied through a VEGA3 TESCAN SEM instrument at room temperature and 10 kV. All samples were coated with gold using an argon plasma metalizer (sputter coater K575X, Edwards Limited, Crawley, UK) to avoid charging.

2.5.2. Transmission Electron Microscopy (TEM)

A JEM-JEOL 1400 TEM instrument was operated at 100 keV to observe the nanocellulose fibers. The nanocellulose suspension was cast in a carbon-coated grid and then directly observed at room temperature.

2.5.3. Thermogravimetric Analysis

All samples were tested using a TGA/DTG 60 system (serial no. C30565000570) to determine the thermal degradation point at each stage. The test was conducted over the temperature range of 30°C–550°C under

a nitrogen atmosphere. The heating rate was maintained at 10°C/min. The weight of all tested samples was 5–7 mg.

2.5.4. Moisture Absorption

All tested samples were cut into 1 cm × 3 cm pieces and dried in a drying oven to a constant weight. The initial (W_o) and final weights (W_t) were recorded. Moisture absorption was examined in a moisture chamber (relative humidity [RH]: 75%) at 25°C. The percent moisture absorption was calculated using the following equation [12]:

$$Moisture\ Absorption = \frac{W_t - W_o}{W_o} \times 100\%. \quad (1)$$

2.5.5. FTIR spectroscopy

FTIR spectroscopy was used to determine the functional groups of all samples. The FTIR spectrum of all tested samples was recorded using a Perkin-Elmer Frontier FTIR instrument with a resolution of 4 cm^{-1}. The scanned wavenumber range was 600–4000 cm^{-1}.

2.5.6. Soil Burial Test

The biodegradation test was conducted by burying the samples in soil that was purchased from PT. Mahesa Mutiara Tani (Kompos Nesia, Bogor, Indonesia). The soil contained organic carbon (34.25%), nitrogen (2%), ferrous (7.9%), pH (7.33), diphosphorus pentoxide (0.62%), other materials (e.g., plastic, glass, gravel = 0.1%), and several colonies in 1 gram of soil (8×10^{14} CFU), and its carbon-to-nitrogen ratio was 17.15%.

Control sample: The sample was cut into 10 mm × 10 mm pieces and then dried in a drying oven at 50°C for 20 h to a constant weight. Then, the sample was subjected to ultrasonic bath treatment for 5 min and dried in an oven at 50°C for 20 h. Subsequently, the sample was weighed using a precision balance (accuracy = 0.001). The weight loss of the control sample was calculated as the difference between W_o before ultrasonic bath treatment and W_t after the treatment.

Buried samples: All samples for the burial test measured 10 mm × 10 mm. The samples were dried in a drying oven at 50°C for 20 h to a constant

weight and weighed using a precision balance to obtain W_o before burial in soil. Then, the test was conducted in a square container (10 cm × 10 cm) containing soil an RH and temperature of 60%–65% and 25°C, respectively. After vibration, biodegradation was observed after 3, 7, and 15 days of burial in soil.

After burial, all samples were collected and cleaned using a paintbrush. Then, the samples were subjected to ultrasonic bath treatment for 5 min to remove the soil that adhered to the biocomposites. The samples were dried in a drying oven at 50°C for 20 h to a constant weight. Afterward, they were weighed using an analytical balance. The percent weight loss of the sample due to soil burial was calculated according to the different weights before and after the burial test. The percent weight loss due to soil was calculated using the following equation:

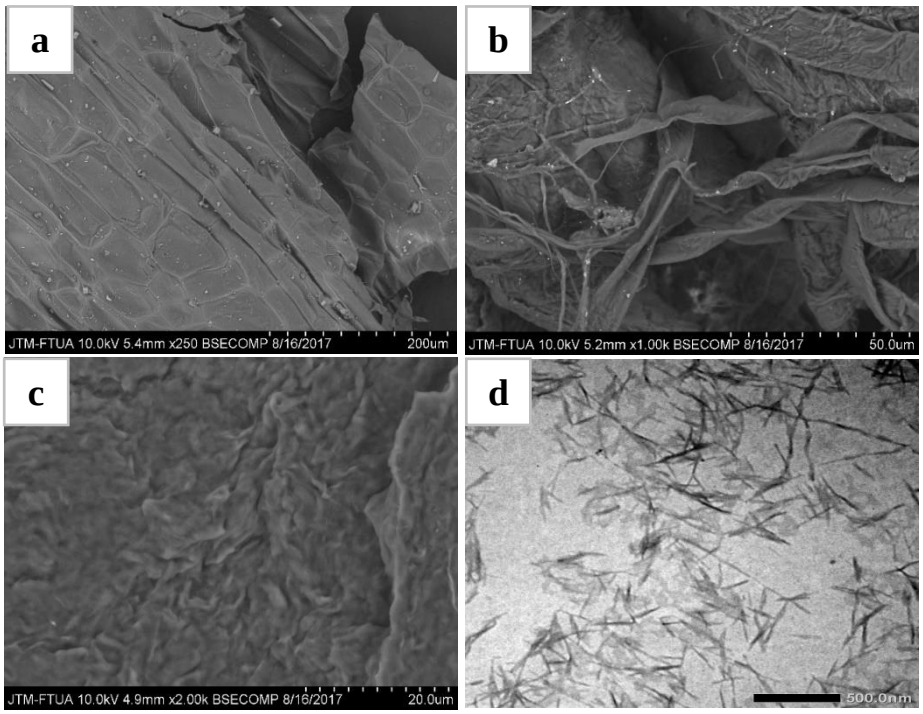
$$W_{degradation} = W_{burial\,test} - W_{control}. \quad (2)$$

3. RESULTS AND DISCUSSION

3.1 Morphological characteristics of untreated and treated fibers

Figure 1 shows the morphological features of water hyacinth fiber before and after treatment. Figure 1a displays the raw water hyacinth fiber (untreated), which had a smooth surface, and some microfibrils were still bound. Untreated fiber contains lignin substances such as waxes and oils [16, 17]. Bleaching successfully destroyed several microfibril bonds, as presented in Figure 1b, because of the broken linkage between lignin and hemicellulose. A similar phenomenon was reported by a previous study [9, 18]. In the acid hydrolysis process (Figure 1c), the fiber was depolymerized into short individual microfibrils with a diameter and length of 2 and 6 microns, respectively. Nanocellulose was obtained via ultrasonication for 1 h. Figure 1d displays the distribution of cellulose fibers in the nanometer range. Cellulose fibers were dispersed homogeneously, as shown in the TEM images (Figure 1d), measuring 15 nm in diameter and 147 nm in length. This outcome is attributable to the high intensity of the power ultrasound, resulting in short cellulose fibers in the nanometer range [19, 20].

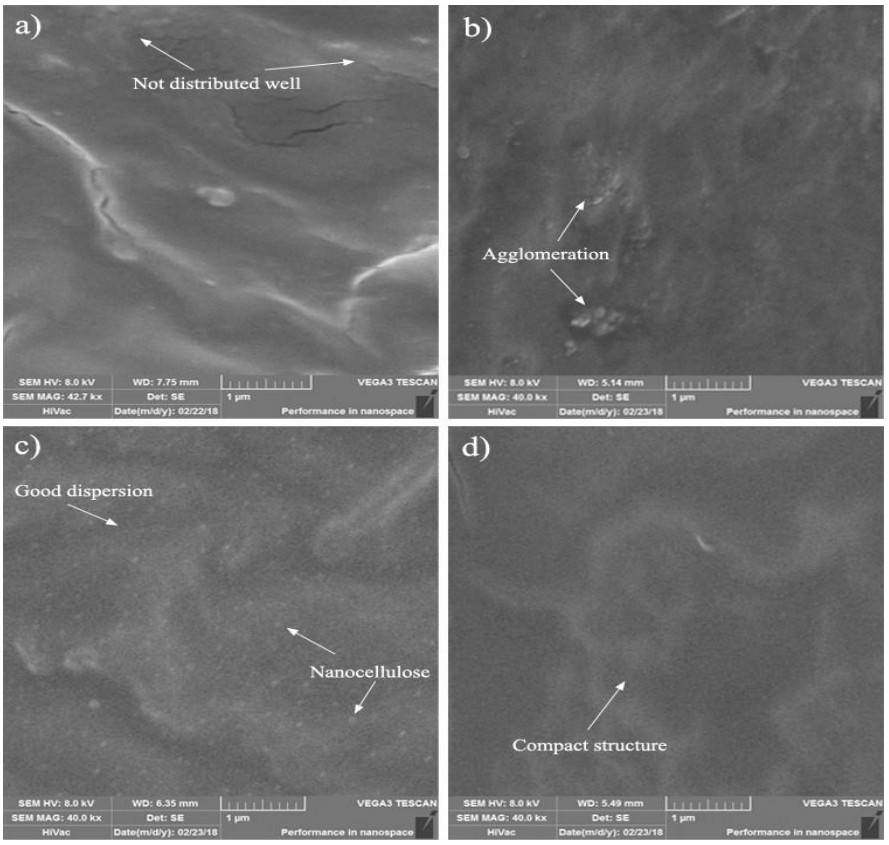
Figure 1. Water hyacinth fiber before and after treatment. (a) Raw fiber (untreated), (b) bleached fiber, (c) fiber subjected to acid hydrolysis; and (d) fiber subjected to ultrasonication.



3.2. Fracture Surface of the Biocomposites

Figure 2 shows the fracture surface of untreated and treated biocomposite films. Figure 2a displays the fracture morphology of the VT 0 min sample (untreated). The cellulose fibers in bengkuang starch were not distributed well. Thus, the mixing of starch and cellulose was not homogeneous during fabrication [21]. This explained the low thermal stability of the untreated biocomposite. A poor distribution of nanocellulose affects thermal stability in terms of interfacial adhesion between the nanocellulose and starch biopolymers. That is, the strong interfacial adhesion between nanocellulose and starch corresponded to the high thermal stability of the biocomposites because a high temperature is required to break the bonds between nanocellulose and starch biopolymers.

The VT 15 min sample also exhibited no significant fracture morphology. The agglomeration phenomenon remains apparent in this sample (Figure 2b). According to a previous report, agglomeration and poor dispersion of cellulose fibers lead to decreased thermal and moisture resistance [22]. The different fracture morphologies are shown in Figures 2c and 2d for the VT 30 min and VT 60 min samples, respectively. In these cases, the samples displayed good dispersion and compact structures. These findings are probably attributable to the kinetic energy from the ultrasonic bath, leading to improved interfacial bonding between the fibers and matrix [12, 13]. This treatment also resulted in good dispersion of the nanocellulose in the matrix and reduced the free OH bonding between the fibers and matrix. This phenomenon was similar to that observed by Asrofi et al. [6] concerning the effect of ultrasonic vibration during processing on



the mechanical properties of a water hyacinth nanofiber cellulose-reinforced thermoplastic starch bionanocomposite.

Figure 2. Fracture surface of all biocomposite samples. Samples were vibrated for (a) 0, (b) 15, (c) 30, or (d) 60 min.

3.3. Thermal Stability

The thermal characteristics of the biocomposite samples with various vibration times were presented using TGA (Figure 3a) and DTG (Figure 3b) curves. Two main degradation areas were found in accordance with the results from previous research [23–25]. The first region of degradation reflected the initial degradation of all biocomposite samples at temperatures of less than 100°C as indicated by the TGA and DTG curves. This result revealed increasing weight loss compared with W_o before testing. The percent weight loss values of the biocomposite in the first region were 11% (VT 0 min), 9% (VT 15 min), 9% (VT 30 min), and 7% (VT 60 min). This phenomenon was attributable to the moisture content in all samples [8, 25]. These results were supported by the DTG curve (Figure 1b), which reveals the presence of small peaks in the temperature range of 50°C–100°C.

The second region (250°C–350°C) involved significant weight loss exceeding 50% for all samples. In this condition, the structures of starch, glycerol, and nanocellulose fibers were degraded [12, 26]. The DTG curve (Figure 1b) exhibits a sharp peak due to the significant weight reduction. The degradation temperatures for all samples in this area was 305°C, 309°C, 311°C, and 318°C for VT 0, VT 15, VT 30, and VT 60 min, respectively. Untreated biocomposite samples (VT 0) min had lower degradation temperatures. This outcome indicated fiber agglomerated and porosity formation in the matrix that resulted in reduced thermal stability [4]. Thermal stability and moisture resistance increased with prolonged ultrasonic vibration.

Conversely, increased vibration time improved the thermal stability of the biocomposite samples. This result was supported by the increase in the degradation temperature of the treated samples compared with that of the untreated sample due to the improved interfacial adhesion between the fibers and matrix as a result of the kinetic energy of the ultrasonic bath. Good adhesion was indicated by the scarcity of free OH between the fibers and matrix [6, 8, 12].

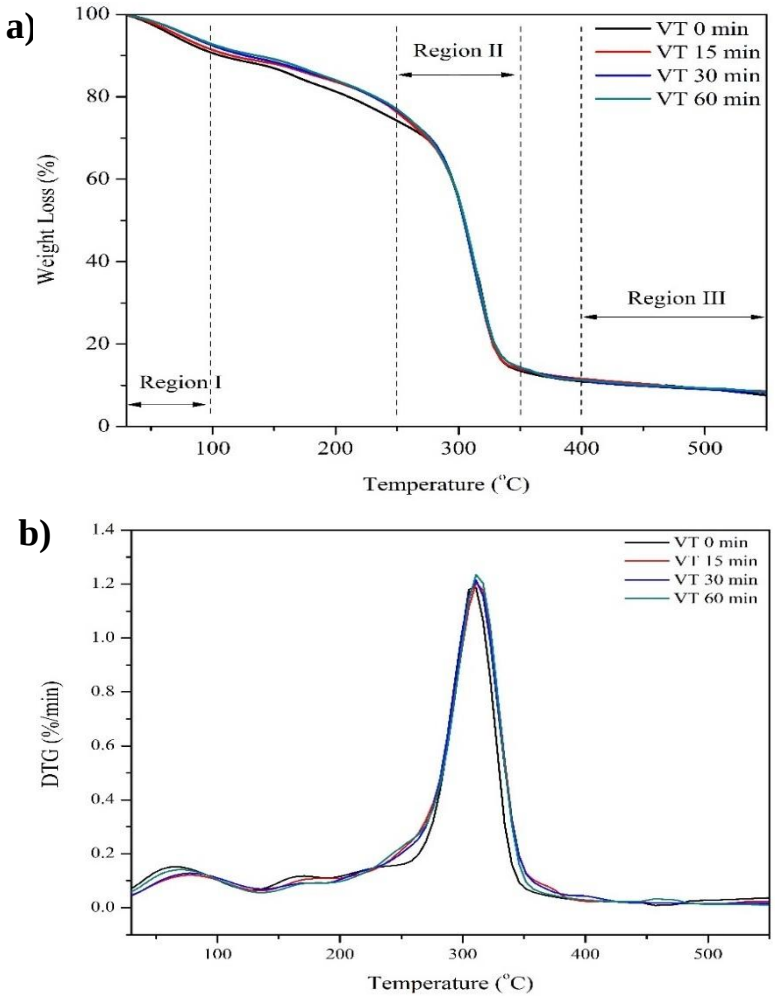


Figure 3. (a) Thermogravimetric analysis and (b) DTG curves of all biocomposite samples

In the third region (>400°C), all samples were completely decomposed, becoming ash [25, 26]. However, the treated samples had higher thermal stability than untreated samples because of the kinetic energy of the ultrasonic bath, which allowed the fibers to disperse evenly in the matrix. Other effects of the bath included good dispersion of the fibers in the

matrix, a compact structure, and improved interfacial bonding between the fibers and matrix. These thermal results are supported by SEM data and are similar to those reported in previous studies [12–13].

3.4. Moisture Absorption

Figure 4 shows the effect of the vibration time on the percent moisture absorption of the biocomposite samples. The test was conducted for 8 h under conditions of RH = 75% and 25°C. All biocomposite samples were saturated in less than 5 h. The percent moisture absorption of the VT 0 min biocomposite sample was 22%. This phenomenon was attributable to the nonhomogeneous dispersion of nanocellulose in the starch matrix. Another reason was the hydrophilic nature of starch and cellulose fibers [13].

These results differed from those of treated biocomposite samples, which exhibited less moisture absorption. For example, in the VT 60 min samples, the percent moisture absorption was 4.5% lower than that of the VT 0 min samples. This outcome occurred because the kinetic energy produced by the ultrasonic bath could reverse fiber agglomeration, homogenously spread cellulose fibers in the starch matrix, and create a strong hydrogen bonding interaction between nanocellulose and the starch biopolymer. Accordingly, it was difficult for water molecules to diffuse into the matrix because of the formation of the tortuous path made by nanocellulose, the high moisture content of which created a barrier effect on the movement of water molecules through the biocomposite [12–13]. This result was supported by the FTIR characterization at a wavenumber of 1600 cm^{-1}, which indicated water absorption by OH groups. Given the chemical similarity between nanocellulose and the starch biopolymers, the interfacial defects were reduced, thereby generating firm resistance to the flow of water molecules.

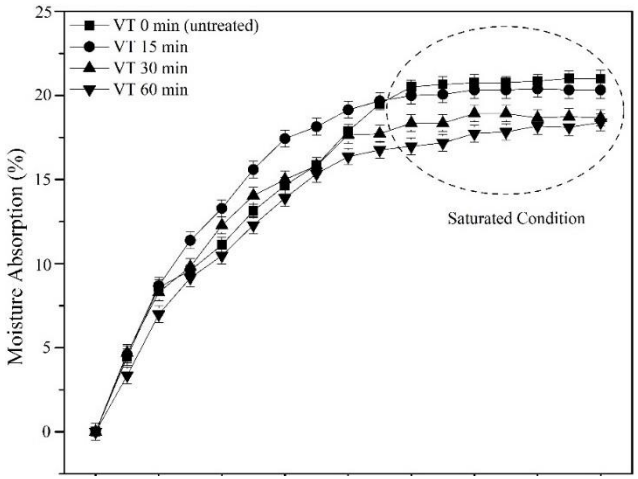


Figure 4. Moisture absorption of all tested biocomposite samples. Samples were subjected to ultrasonication for 0, 15, 30, or 60 min. VT: vibration time.

3.5. Functional group analysis

Figure 5 shows the FTIR spectra of untreated and treated biocomposites. Three main peaks occurred at different wavenumbers (Figure 5), namely 3000 (O-H stretching), 2900 (C-H stretching), and 1600 cm^{-1} (water absorption by OH groups) [27, 28].

O-H stretching appeared at 3000 cm^{-1}, and the biocomposite displayed a shift to higher wavenumbers. This result indicated an interaction between the matrix and fibers that generated new hydrogen bonds [28]. The transmittance values of the untreated and treated biocomposites varied. Long vibration times resulted in high transmittance values (Figure 5) because of the greater OH stretching. Good hydrogen bonding was noted between the matrix and fibers (few occurrences of free OH) [12], thereby producing satisfactory tensile strength due to the good structure formation of OH molecules (crystalline) [6, 12].

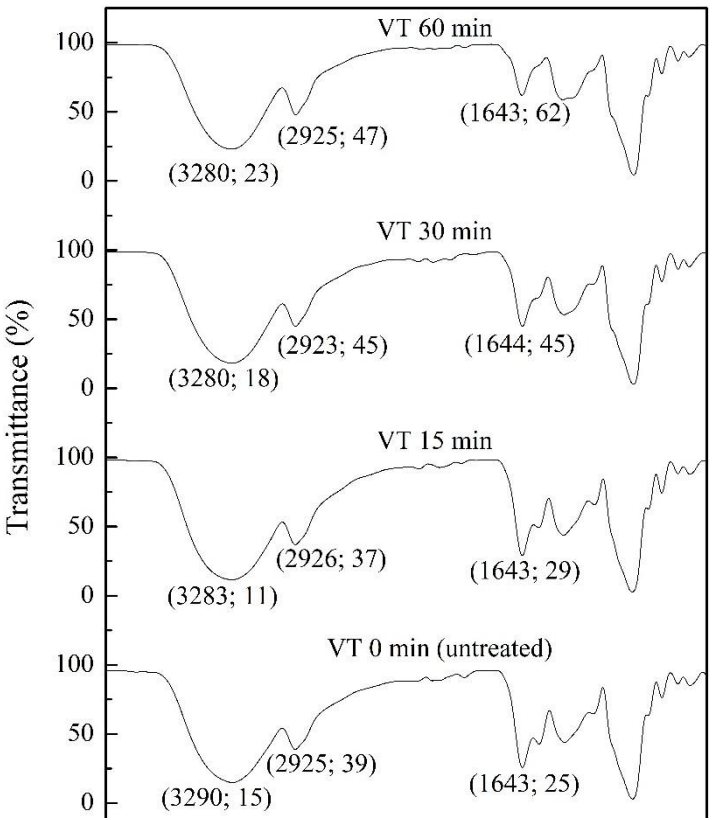


Figure 5. Functional groups of all tested biocomposite samples. VT: vibration time.

In addition, C-H stretching appeared at approximately 2900 cm^{-1} in all biocomposite samples. Thus, all samples contained saturated aliphatic components, as reported by previous researchers [29]. Another phenomenon also appeared around 1600 cm^{-1}. In this area, OH water absorption appears in each biocomposite sample. The transmittance value increases with the duration of vibration. The transmittance values of the VT 0, VT 15, VT 30, and VT 60 min biocomposite samples were 25%, 29%, 45%, and 62%, respectively. These results suggest that untreated biocomposite samples exhibit greater water absorption than treated samples [4, 28]. This outcome was supported by the moisture absorption test results, in which the VT 0 min samples had the highest percent moisture absorption. However, treated biocomposite samples displayed lower moisture absorption. The kinetic energy of the ultrasonic bath could degrade the agglomerated nanocellulose fiber and spread them evenly in the matrix [12]. This phenomenon makes it difficult for water molecules to diffuse into the matrix. A similar outcome was reported by previous research [6, 12].

3.6. Soil Biodegradation

Investigating soil biodegradation behavior is crucial for utilizing biocomposites in the environment. Soil biodegradation involves the degradation of materials by the action of microorganisms, fungi, bacteria, or other organisms that live in soil. Figure 6 shows the effect of the vibration time on the biodegradation rates of all biocomposite samples. Compared with treated samples, untreated biocomposites exhibited greater weight loss. The percent weight loss on day 3 for untreated samples was 9.55%,

and the values on days 7 and 15 were 30.06% and 31.21%, respectively. This outcome was supported by the results presented in Figures 7a and 7b, in which the morphological structure depicts a large hole with an irregular configuration. This phenomenon may be attributable to fiber clumping in the matrix [8].

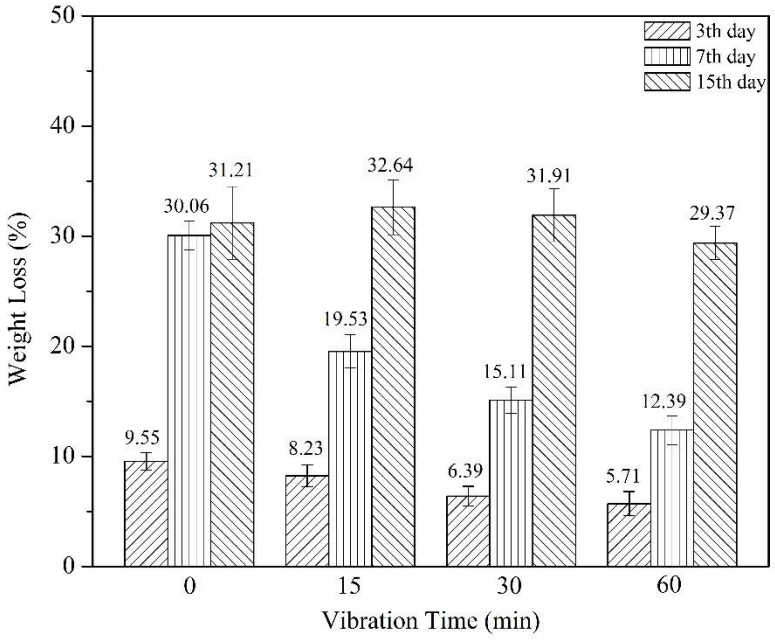


Figure 6. Biodegradation rates of all tested biocomposite samples

These values differed from those of VT 60 min samples. In these samples, the percent weight loss values were 5.71% (day 3), 12.39% (day 7), and 29.37% (day 15). Thus, VT 60 min samples had a slower degradation rate than the untreated samples. This phenomenon was attributable to the kinetic energy from the ultrasonic bath, which distributed the fibers homogeneously within the matrix [6]. The good fiber distribution in the matrix affected moisture, microorganisms, and other elements such that

their diffusion into the matrix was difficult, as reported by previous research [14, 15, 30–32]. These results were supported by the SEM findings (Figure 7c and 7d), in which the biocomposite structure was compact and less porous. The vibration time also decreased the biodegradation rate. These phenomena may be correlated with the water absorption properties of the biocomposite film, in which untreated film tends to absorb more water than treated film, thereby making the former more prone to attacks by microorganisms, which can attack the starch biopolymer in the presence of a water medium.

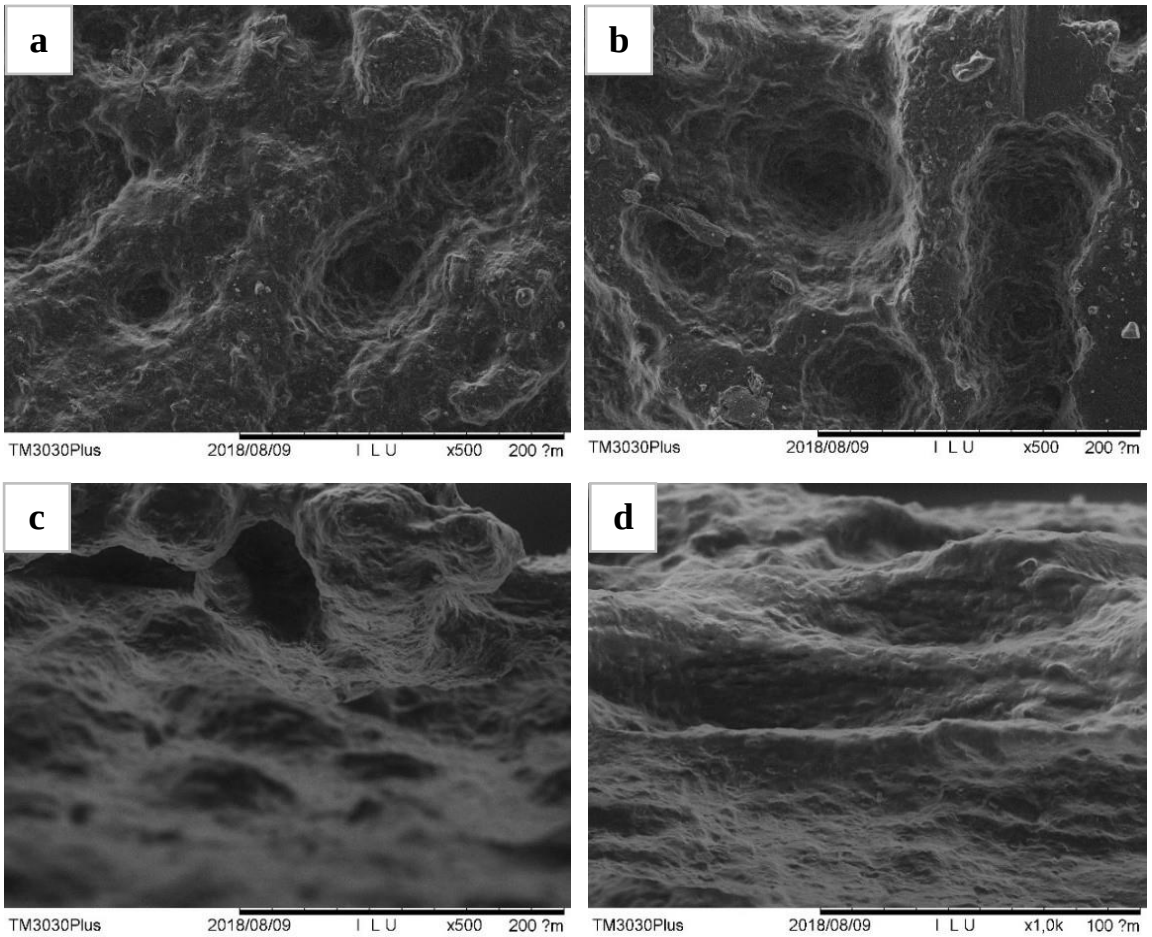


Figure 7. Morphological characteristics of biocomposite samples after biodegradation in soil. (a) Untreated (day 3), (b) untreated (day 15), (c) vibrated for 60 min (day 3), and (d) vibrated for 60 min (day 15).

4. CONCLUSION

Thermal stability and moisture resistance were successfully improved by vibration treatment using an ultrasonic bath instrument. Kinetic energy from the ultrasonic bath reduced the free OH bonding between the fiber and matrix. The best results were obtained for VT 60 min biocomposite samples. Increasing the vibration time also reduced the degradation rate of biocomposites in the soil. The soil burial test revealed that the vibrated biocomposites had slower biodegradation rates than the samples. The properties of the biocomposites suggest their potential application as environmentally friendly plastics for food packaging. The potential application of this biocomposite is food packaging, especially as packaging bags.

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CONFLICTS OF INTEREST

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CONFLICTS OF INTEREST

The authors declare no conflict of interest regarding the publication of this.

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3. Publish

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Original Article

Effect of sonication time on the thermal stability, moisture absorption, and biodegradation of water hyacinth (*Eichhornia crassipes*) nanocellulose-filled bengkuang (*Pachyrhizus erosus*) starch biocomposites

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ABSTRACT