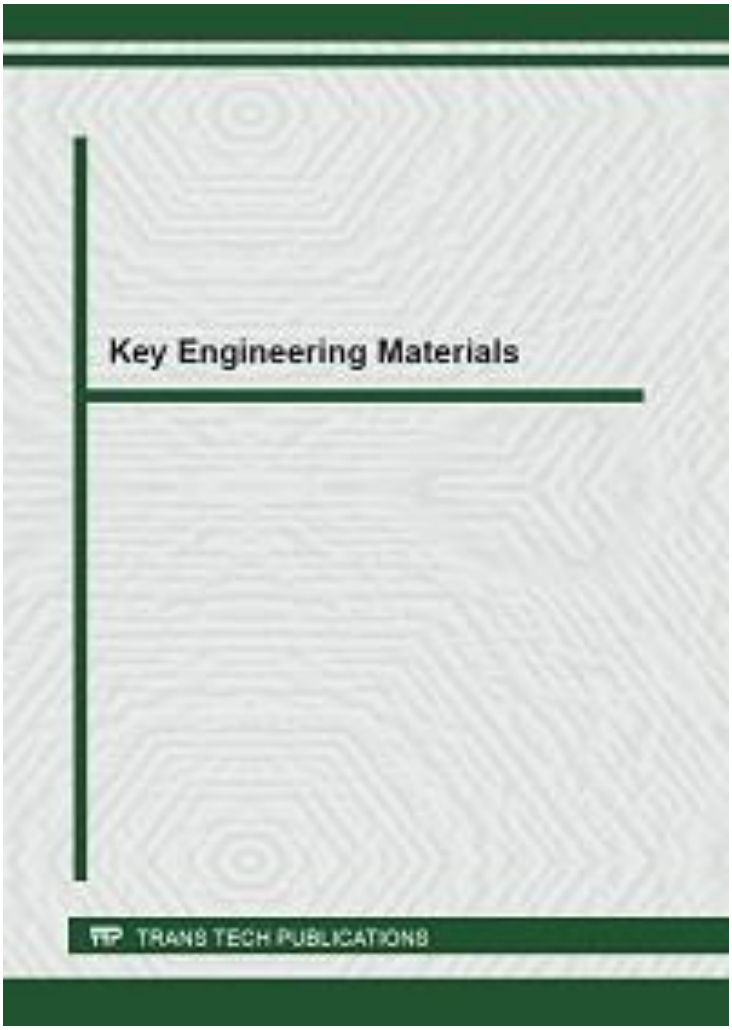
Paper 9 Not open access :
Improvement of Biocomposite Properties Based Tapioca Starch and Sugarcane Bagasse Cellulose Nanofibers

Silahkan klik google drive utk full access: https://drive.google.com/file/d/1-AlQvp3l_8SBfXq4KYGbE-5NTOoaIaj1/view?usp=sharing



Improvement of Biocomposite Properties Based Tapioca Starch and Sugarcane Bagasse Cellulose Nanofibers

Mochamad Asrofi1,A , Sujito,B, Edi Syafri, S.M. Sapuan And R.A. Ilyas

Key Engineering Materials - Editorial Board

ISSN: 1662-9795

Details | Volumes | Editorial Board

Key Engineering Materials

COUNTRY

Switzerland

Universities and research institutions in Switzerland

Media Ranking in Switzerland

SUBJECT AREA AND CATEGORY

Engineering
- Mechanical Engineering
- Mechanics of Materials

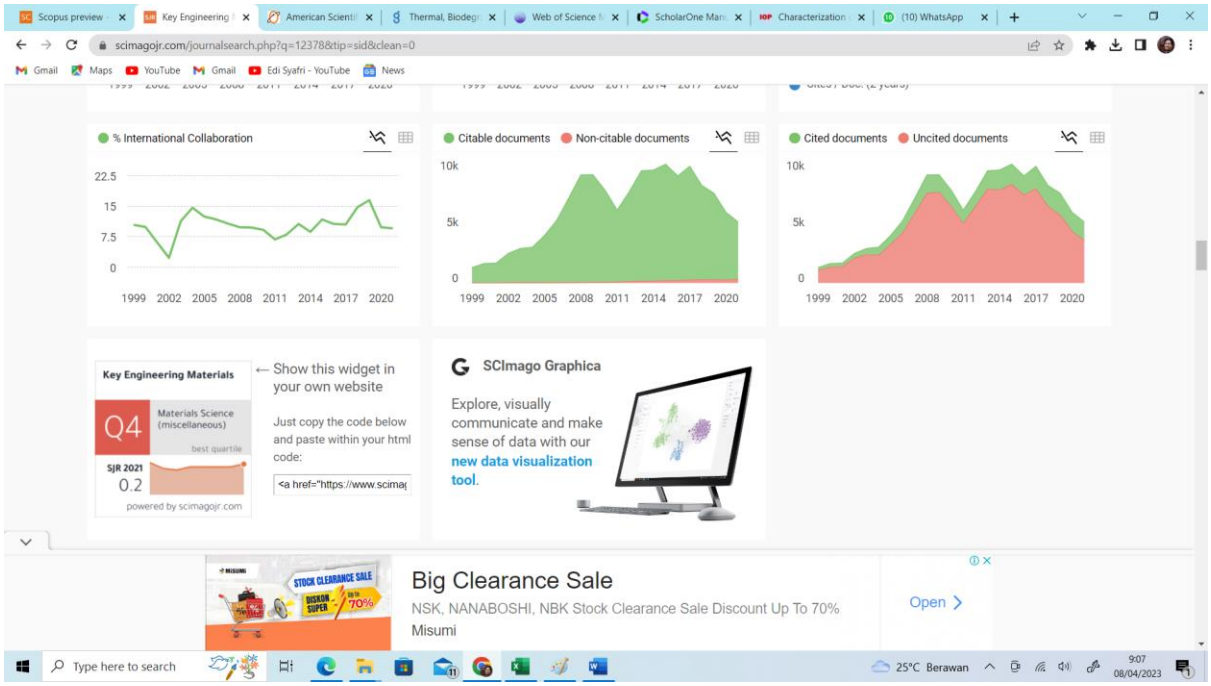
Materials Science
- Materials Science (miscellaneous)

PUBLISHER

Trans Tech Publications

H-INDEX

55


scimagojr.com/journalsearch.php?q=12378&tip=sid&clean=0
% International Collaboration
22.5
15
7.5
0
1999 2002 2005 2008 2011 2014 2017 2020
Citable documents
Non-citable documents
10k
5k
0
1999 2002 2005 2008 2011 2014 2017 2020
Cited documents
Uncited documents
10k
5k
0
1999 2002 2005 2008 2011 2014 2017 2020
Key Engineering Materials
Q4
Materials Science (miscellaneous)
best quartile
SJR 2021
0.2
powered by scimagojr.com
Show this widget in your own website
Just copy the code below and paste within your html code:
<a href="https://www.scimag
SCImago Graphica
Explore, visually communicate and make sense of data with our new data visualization tool.
STOCK CLEARANCE SALE
DISKON SUPER
70%
Big Clearance Sale
NSK, NANABOSHI, NBK Stock Clearance Sale Discount Up To 70%
Misumi
Open
Type here to search
25°C Berawan
9:07
08/04/2023

List of Contents
Volume 849

Waste and Biomass Application

ISBN(softcover): 978-3-0357-1698-6 ISBN(eBook): 978-3-0357-3698-4

Table of Contents

Chapter 2: Production of Materials and Chemicals from Waste and Biomass

Improvement of Biocomposite Properties Based Tapioca Starch and Sugarcane Bagasse Cellulose Nanofibers

Article *in* Key Engineering Materials · June 2020

DOI: 10.4028/www.scientific.net/KEM.849.96

CITATIONS
3

READS
103

5 authors, including:

Mochamad Asrofi
Universitas Jember
44 PUBLICATIONS **555** CITATIONS
SEE PROFILE

Sujito M. Sofyan
Universitas Jember
10 PUBLICATIONS **10** CITATIONS
SEE PROFILE

Edi Syafri
Payakumbuh State Polytechnic of Agriculture
34 PUBLICATIONS **438** CITATIONS
SEE PROFILE

S. M. Sapuan
Universiti Putra Malaysia
923 PUBLICATIONS **16,996** CITATIONS
SEE PROFILE

Key Engineering Materials
ISSN: 1662-9795, Vol. 849, pp 96-101
doi:10.4028/www.scientific.net/KEM.849.96

Submitted: 2019-10-11
Revised: 2020-01-15
Accepted: 2020-01-15
Online: 2020-06-24

Improvement of Biocomposite Properties Based Tapioca Starch and Sugarcane Bagasse Cellulose Nanofibers

MOCHAMAD Asrofi[1,a*], SUJITO[2,b], EDI Syafri[3,c], S.M. Sapuan[4,d] and R.A. Ilyas[4,e]

[1]Laboratory of Material Testing, Department of Mechanical Engineering, University of Jember, Kampus Tegalboto, Jember 68121, East Java, Indonesia

[2]Department of Physics, University of Jember, Jl. Kalimantan No. 37, Jember, 68121, East Java, Indonesia

[3]Department of Agricultural Technology, Agricultural Polytechnic, Payakumbuh, West Sumatra 26271, Indonesia

[4]Advanced Engineering Materials and Composites Research Centre, Department of Mechanical and Manufacturing Engineering, Universiti Putra Malaysia, 43400 UPM Serdang, Selangor, Malaysia

[a]asrofi.teknik@unej.ac.id, [b]sujito.fmipa@unej.ac.id, [c]edisyafri11@gmail.com, [d]sapuan@upm.edu.my, [e]ahmadilyasrushdan@yahoo.com

Keywords: Sugarcane Bagasse Fiber, Cellulose Nanofibers, Tapioca Starch, Biocomposite, Tensile Properties.

Abstract. Biocomposite based tapioca starch (TS) and sugarcane bagasse cellulose nanofibers (SBCN) was made through casting method. SBCN was prepared by chemical and ultrasonication process. It was successfully displayed by transmission electron microscope (TEM) in range 20 - 45 nm. Meanwhile, particle size analysis (PSA) also supported the distribution diameter of SBCN for 59.75 ± 10.84 nm. SBCN and glycerol were used as reinforcement and plasticizer, respectively. The amount concentration of SBCN was varied from 0 to 8 wt%. Biocomposite was characterized by using scanning electron microscopy (SEM) and tensile test. SEM image displays SBCN is in good interfacial bonding with the matrix. The highest tensile strength of biocomposite was in TS/4SBCN sample for 20.84 MPa. These results showed that SBCN fiber become potential candidate as reinforcement in biocomposite application.

Introduction

The development of biodegradable materials from renewable sources is increasing due to environmental awareness. Among of natural polymers, starch is considered as one of the most promising candidates because its advantages such as, availability, low cost and good performance [1,2]. Tapioca (*Manihot esculenta*) is an important source of starch in several countries such as Thailand, Malaysia, Indonesia and some African regions. In addition, using of tapioca starch for bioplastics composite has been developed by many previous researchers [3-5]. In its application, starch has several limitations such as low tensile strength and moisture resistance. One solution to overcome this limitation is the addition of with organic and inorganic fillers in starch matrix [6, 7].

Cellulose nano fibers (CNF) is a new type of filler which shows high efficiency in improving the physical and mechanical properties of starch-based biocomposite. Nano particle-sized fillers have good compatibility with matrix due to the high contact surface area [8]. One of the fiber plants which has a high cellulose content is sugarcane bagasse. Sugarcane bagasse fiber has high α-cellulose content approximately 85%. Other advantages are good mechanical properties, low density, low cost and abundant in earth. Therefore, sugarcane bagasse fiber has potential to reinforce starch biocomposite [9].

Several previous studies have explained that sugarcane bagasse cellulose has a good bond with the starch matrix [9, 10]. This is indicated by the hydroxyl bond formation between cellulose and starch resulting in improvement of mechanical properties [11]. Meanwhile, the fiber size of

sugarcane bagasse is also play important factor due to its function in filling the void fraction in starch matrix. Previous studies reported that nano-sized cellulose sugarcane bagasse was effective in increasing the properties of biocomposite. Nano size fiber can spread evenly into the starch matrix. Even distribution makes less voids formation [8, 12].

In this study, the synthesis of SBCN was carried out by the chemical-ultrasonic method. The SBCN was added into tapioca starch matrix as reinforcement. The addition of SBCN to the matrix is expected to improve the tensile properties of biocomposite. According to the best our review, there is no research reported about SBCN reinforced tapioca starch matrix. We state that the research about sugarcane bagasse cellulose fiber in nanoscale is limited information. The synthesis of SBCN was produced by chemical and ultrasonication method namely pulping, bleaching, sulfuric acid hydrolysis and ultrasonication. The characterization of SBCN was carried out by using particle size analyzer (PSA) and transmission electron microscopy (TEM), while biocomposites were characterized by using scanning electron microscopy (SEM) and tensile test.

Materials and Methods

Materials. Sugarcane bagasse was obtained from local sugarcane ice seller at Jember, Indonesia. Sodium hydroxide (98% NaOH), potassium hydroxide (KOH), sodium chlorite ($NaClO_2$), acetic acid (CH_3COOH), sulfuric acid (H_2SO_4) and glycerol were purchased from CV. Aneka Kimia located at Jember, Indonesia. Tapioca starch was supplied by Laboratory of Material Testing, Department of Mechanical Engineering, University of Jember, Jember, Indonesia.

Preparation of sugarcane bagasse cellulose nanofibers (SBCN). The overall isolation of SBCN was in accordance previous method and modified in hydrolysis process [13, 14]. Hydrolysis was carried out before the ultrasonication process. Hydrolysis process used 30% H_2SO_4 for 60 min with ratio fiber and solution (1 : 8.75). After hydrolysis, sugar cane bagasse fiber was washed by distilled water until neutral pH. Then, it was ultrasonicated by using sonicator (Ultrasonic 750W) at 70 ^oC for 150 min to obtain SBCN.

Fabrication TS/SBCN biocomposites. The composition of biocomposite from SBCN and tapioca starch was presented in Table 1. Tapioca starch was dissolved in distilled water (% w/v) and SBCN (% w/w based tapioca starch) with different concentrations. Glycerol as a plasticizer (30% w/w from tapioca starch) was added in mixing solution. Then, the biocomposite solution was heated at 65-75^oC with constant stirring (200 rpm) until gelatinized, poured into an acrylic mold (11 cm x 9.5 cm x 0.3 cm). To remove air bubbles, the mold was placed in an ultrasonic batch for 15 min [15, 16]. After that, it was dried in drying oven at 40^oC for 17 h. Biocomposite was stored in the desiccator for 24 h before characterization.

Table 1. The composition of the biocomposite from tapioca starch reinforced SBCN

Biocomposites code	Glycerol (w/w dry starch basis)	TS, w/40 ml aqudest	SBCN (% dry starch basis)
TS (Control)	0.3	2	0
TS/2SBCN	0.3	2	2
TS/4SBCN	0.3	2	4
TS/8SBCN	0.3	2	8

**This composition is similar with previous report but difference in the addition of % fiber content and glycerol volume [17].*

SEM and TEM. The surface morphology of biocomposite was observed using SEM (Model JSM 6510 LA from JEOL) with a voltage and flow at probe for 15 kV and 8 mA, respectively. The tested sample was placed on the SEM stub sample. The preparation sample was previously coated with carbon followed by coating with gold to reduce electron charge. SEM images were enlarged to

get image clarity. Furthermore, the morphology of SBCN was observed using TEM-Tecnai T20 at 200 kV for microstructure observation. The suspension of SBCN after ultrasonication process was dripped a few drops over the holley carbon grid and dried at room temperature for 4 h. The dried SBCN solution was directly observed by TEM instrument.

Tensile strength. The tensile strength of biocomposite sample was determined using a tensile machine at room temperature conditions of 25ºC. Tensile specimen was formed according to the American Standard Testing Material (ASTM) D-638 type IV. The test was maintained on crosshead speed of 2 mm/min using a load cell of 5 kN load. Five samples were tested for each biocomposite variation as stated in previous report [17].

Results and Discussions

Morphological of SBCN. PSA and TEM images of SBCN suspension are shown in Figures 1a and 1b, respectively. The average size of SBCN produced by the chemical-ultrasonication method is 59.75 ± 10.84 nm. This SBCN diameter is smaller in size compared to the hydrolysis-homogenization process which carried out by previous report about isolation of nanocellulose from sugarcane bagasse for 95 nm [18]. This proved that the hydrolysis treatment with H_2SO_4 and ultrasonication had successfully broken the hydrogen bonds between cellulose [13,14]. This is supported by the SBCN morphology by TEM (Figure 1b) which shows SBCN are disintegrated and spread evenly.

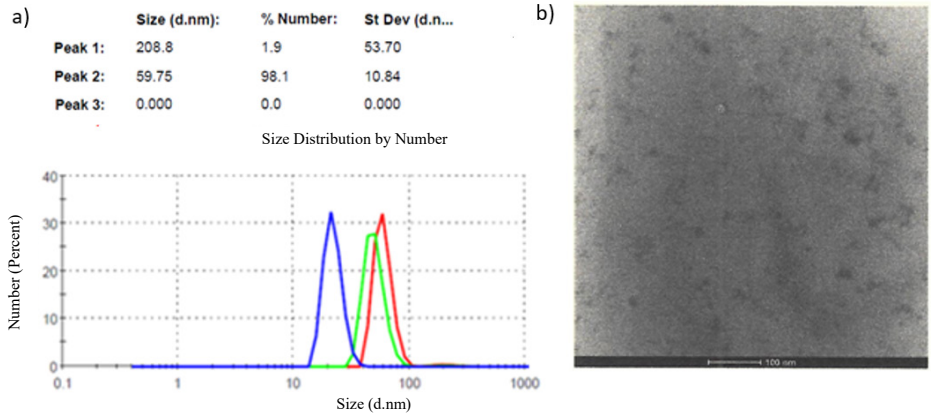


Fig. 1. SBCN distribution by different characterization: a) PSA and b) TEM

According to previous researchers, the ultrasonication process was very effective for breaking the hydrogen cellulose chain in the liquid medium. The ultrasonication emits ultrasonic waves and form cavitation in a liquid medium. The cavitation formed will collide with each other to break the bond. However, this process has a disadvantage such as high-power electricity consumption [15,19].

Morphology SEM of TS/SBCN Biocomposite. Fig. 2 shows the fracture surface morphology of a TS/SBCN biocomposite. Figure 2a displays the control sample (pure TS film). It can be seen that fracture surface looks smoother than biocomposite samples. This phenomenon is caused by good mixing between glycerol and tapioca starch during gelatinization process [8]. Good mixing and dispersion able to make homogeneous film. A similar phenomenon was also reported by previous studies of films from yam bean starch [6, 12]. The addition of SBCN in the tapioca starch matrix displays a rougher surface. The addition of 2%SBCN into TS matrix (Fig. 2b) showed less dispersion of SBCN particles than 4% and 8% SBCN in TS matrix. This fracture is also accompanied by cracks in various parts. This is due to the bad preparation of biocomposite samples [20].

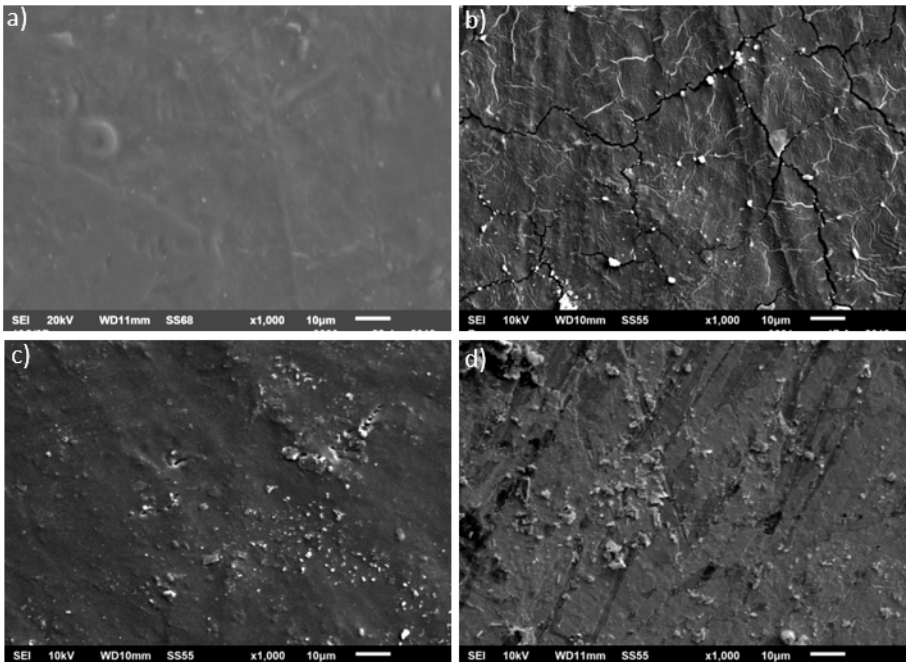


Fig. 2. Fracture morphology by SEM image: a) 0% (control), b) 2%, c) 4%, d) 8% of SBCN

Meanwhile, the addition of 4% SBCN in TS matrix (Fig. 2c) shows a more homogeneous surface and it has denser structure. The SBCN particles looks evenly dispersed in various parts of the matrix. This results in a good bond between the fiber and matrix resulting in higher tensile strength [16]. Different phenomena are shown by 8% SBCN in the TS matrix (Fig. 2d). This sample shows porosity in several parts. In addition, clot formations are formed in these samples. This is due to the ineffective transfer of stress from fiber to matrix [15, 21]. This factor affected in decreasing mechanical properties. This result is also supported by the tensile strength which shows a decreasing up to 200% compared to the addition of 4% SBCN in TS matrix.

Tensile strength and strain of TS/SBCN biocomposites. The tensile strength and tensile strain of TS/SBCN biomposites are shown by Fig. 3a and 3b, respectively.

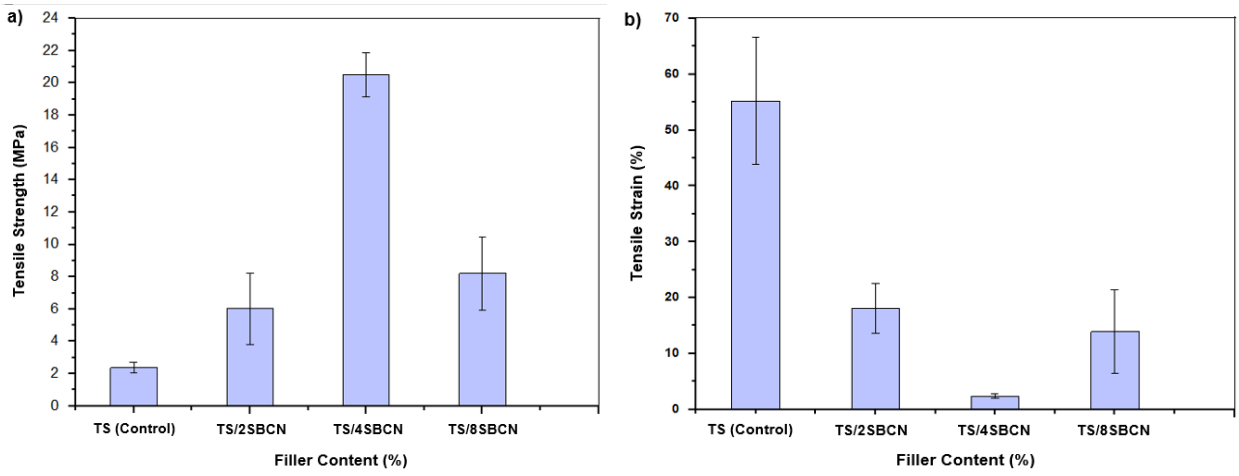


Fig. 3. Tensile properties of all samples tested: a) tensile strength and b) tensile strain of all biocomposites tested

It can be seen that the tensile strength increased significantly from 2.37 ± 0.34 MPa to 20.51 ± 1.37 MPa with the addition of 4% SBCN in starch matrix (Figure 3a). This tensile strength is higher than the previous research as conducted by previous report for 6.68 MPa (10% fiber in starch matrix) [22]. Conversely, there was a decrease in tensile strain 4%SBCN biocomposites from 55.27% (control) to 2.37% (Fig. 3b). This indicates that the SBCN particles are distributed evenly

in tapioca starch matrix for 4% SBCN samples. In addition, good adhesion bonding between matrix and fiber is also become supporting factor in increasing tensile strength [12, 22]. This result was supported by SEM imaging (Fig. 2c). The addition of SBCN to the TS matrix makes the sample become brittle (strain reduced). It is affected by the present of SBCN in TS matrix which results in the prevention mobility of starch chain. Similar results were reported by previous studies [15].

Conclusion

SBCN was successfully synthesized by chemical-ultrasonication method with a diameter of 59.75 ± 10.84 nm. This SBCN has potential used as reinforcement in biocomposite based starch due to its nanosized fiber. The SBCN was found in good dispersion in biocomposite based tapioca matrix. Beside that, the mixing SBCN and tapioca starch resulting in good adhesion bonding. This is affected in highest tensile strength of 4% SBCN in TS matrix for 20.84 MPa. This improvement in biocomposite properties proves that SBCN might have potential as a reinforcement in biopolymer matrix-based packaging material.

Acknowledgements

This research was funded and supported by Institute of Research and Community Service, University of Jember for beginner lecturer research scheme with project number: 3107/UN25.3.1/LT/2019.

References

[1] D.R. Lu, C.M. Xiao, & S.J. Xu, Starch-based completely biodegradable polymer materials, Express Polym. Lett. 3 (2009) 366-375.

[2] R.A. Ilyas, S.M. Sapuan, M.R. Ishak, & E.S. Zainudin, Development and characterization of sugar palm nanocrystalline cellulose reinforced sugar palm starch bionanocomposites, Carbohydr. Polym. 202 (2018) 186-202.

[3] S. Chiarakorn, C.K. Permpoonwiwat, & P. Nanthachatchavankul, Cost benefit analysis of bioplastic production in thailand. Econ. Public Policy J. 3 (2012) 56-85.

[4] E. Syafri, A. Kasim, H. Abral, G.T. Sulungbudi, M.R. Sanjay, & N.H. Sari, Synthesis and characterization of cellulose nanofibers (CNF) ramie reinforced cassava starch hybrid composites. Int. J. Biol. Macromol. 120 (2018) 578-586.

[5] O.O Oladayo, O.S. Joseph, & W. Oluwasegun, Physicochemical properties of cassava starch and starch-keratin prepared biofilm. Songklanakarin J. Sci. Technol. 38 (2016) 349-355.

[6] S. Mali, M.V.E. Grossmann, M.A. García, M.N. Martino, & N.E. Zaritzky, Mechanical and thermal properties of yam starch films. Food Hydrocolloid. 19 (2005) 157-164.

[7] S. Wahono, A. Irwan, E. Syafri, & M. Asrofi, Preparation and characterization of ramie cellulose nanofibers/$CaCO_3$ unsaturated polyester resin composites. ARPN J. Eng. Appl. Sci. 13 (2018) 746-751.

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

[9] A. Mandal & D. Chakrabarty, Isolation of nanocellulose from waste sugarcane bagasse (SCB) and its characterization. Carbohydr. Polym. 86 (2011) 1291-1299.

[10] K. Saelee, N. Yingkamhaeng, T. Nimchua, & P. Sukyai, An environmentally friendly xylanase-assisted pretreatment for cellulose nanofibrils isolation from sugarcane bagasse by high-pressure homogenization. Ind. Crop. Prod. 82 (2016) 149-160.

[11] W.N. Gilfillan, D.M. Nguyen, P.A. Sopade, & W.O. Doherty, Preparation and characterisation of composites from starch and sugar cane fibre. Ind. Crop. Prod. 40 (2012) 45-54.

[12] M. Asrofi, H. Abral, A. Kasim, A. Pratoto, M. Mahardika, & F. Hafizulhaq, Characterization of the sonicated yam bean starch bionanocomposites reinforced by nanocellulose water hyacinth fiber (Whf): The effect of various fiber loading. J. Eng. Sci. Technol. 13 (2018) 2700-2715.

[13] E. Syafri, A. Kasim, H. Abral, & A. Asben, Cellulose nanofibers isolation and characterization from ramie using a chemical-ultrasonic treatment. J. Nat. Fiber. (2018) 1-11.

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

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

[16] M. Asrofi, H. Abral, Y.K. Putra, S.M. Sapuan, & H.J. Kim, Effect of duration of sonication during gelatinization on properties of tapioca starch water hyacinth fiber biocomposite. Int. J. Biol. Macromol. 108 (2018) 167-176.

[17] E. Syafri, S. Wahono, A. Irwan, M. Asrofi, N.H. Sari, & A. Fudholi, Characterization and properties of cellulose microfibers from water hyacinth filled sago starch biocomposites. Int. J. Biol. Macromol. 137 (2019) 119-125.

[18] W.T. Wulandari, A. Rochliadi, & I.M. Arcana, Nanocellulose prepared by acid hydrolysis of isolated cellulose from sugarcane bagasse. IOP Conf. Ser. Mater. Sci. Eng. 107 (2016) p. 012045

[19] W. Chen, H. Yu, Y. Liu, P. Chen, M. Zhang, & Y. Hai, Individualization of cellulose nanofibers from wood using high-intensity ultrasonication combined with chemical pretreatments. Carbohydr. Polym. 83 (2011) 1804-1811.

[20] M.B. Agustin, B. Ahmmad, E.R.P. De Leon, J.L. Buenaobra, J.R. Salazar, & F. Hirose, Starch-based biocomposite films reinforced with cellulose nanocrystals from garlic stalks. Polym. Composite. 34 (2013) 1325-1332.

[21] J.P. López, P. Mutjé, A.J.F.D. Carvalho, A.A.D.S. Curvelo, & J. Gironès, Newspaper fiber-reinforced thermoplastic starch biocomposites obtained by melt processing: evaluation of the mechanical, thermal and water sorption properties. Ind. Crop. Prod. 44 (2013) 300-305.

[22] H. Abral, M.H. Dalimunthe, J. Hartono, R.P. Efendi, M. Asrofi, E. Sugiarti, S.M. Sapuan, J.W. Park, & H.J. Kim, Characterization of tapioca starch biopolymer composites reinforced with micro scale water hyacinth fibers. Starch-Stärke 70 (2018) 1700287.