UNIVERSITI PUTRA MALAYSIA

EXTRACTION AND CHARACTERIZATION OF CELLULOSE NANOCRYSTALS FROM TEA LEAF WASTE FIBERS AS A FILLER IN POLY(LACTIC ACID) BIO-NANOCOMPOSITES

NUR HAYATI BINTI ABDUL RAHMAN

FS 2018 96

EXTRACTION AND CHARACTERIZATION OF CELLULOSE NANOCRYSTALS FROM TEA LEAF WASTE FIBERS AS A FILLER IN POLY(LACTIC ACID) BIO-NANOCOMPOSITES

By

NUR HAYATI BINTI ABDUL RAHMAN

Thesis Submitted to the School of Graduate Studies, Universiti Putra Malaysia, in Fulfilment of the Requirements for the Degree of Master of Science

September 2018

Abstract of thesis presented to the Senate of Universiti Putra Malaysia in fulfilment of the requirement for the degree of Master of Science

EXTRACTION AND CHARACTERIZATION OF CELLULOSE NANOCRYSTALS FROM TEA LEAF WASTE FIBERS AS A FILLER IN POLY(LACTIC ACID) BIO-NANOCOMPOSITES

By

NUR HAYATI BINTI ABDUL RAHMAN

September 2018

Chairman: Nor Azowa binti Ibrahim, PhD
Faculty: Science

Poly(lactic acid) (PLA) is known to be a very useful material in replacing the petrochemical-based polymer in the packaging sector due to its biodegradability and good mechanical characteristics. Despite the impressive behaviors of PLA, the low brittleness of itself has limited the usage of this material. Thus, cellulose nanocrystals (CNCs) was extracted from tea leaf wastes fibers (TLWF) to incorporate into PLA, enhancing the performance of polymer nanocomposites while keeping the environment safe.

This research was conducted to explore the utilization of TLWF as a source for the production of CNCs and its uses as a filler on PLA. TLWF was first treated with alkaline, followed by bleaching before being hydrolyzed with concentrated sulfuric acid. The materials attained after each step of treatments were characterized. From Fourier transform infrared spectroscopy (FTIR), the peak at 1716 represents C=O stretching disappeared in the spectra after the alkaline and bleaching treatments indicated that hemicellulose and lignin were almost discarded from the fiber. The reduction intensity of the absorption band at 1236 cm^{-1}which accredited to the C–O stretching vibration of the lignin and xylan occurred due to the decreasing of lignin and a small hemicellulose contents from TLWF. Meanwhile, the thermal stability of CNCs was decreased due to the replacement of hydroxyl groups by sulfate groups during hydrolysis. The scanning electron microscopy (SEM) showed the defibrillation of fibers occurred after the treatments, increasing the susceptibility of fiber for acid hydrolysis. The crystallinity index of fiber also increased from 41.5% to 83.1% and from transmission electron microscopy (TEM), the rod-like shaped CNCs with an average diameter of 7.97 ± 1.09 nm was revealed.

Then, the resultant CNCs were used as filler in PLA bio-nanocomposites *via* solvent casting method. The different contents of CNCs between 0.5 to 5 wt % were filled. The mechanical properties of films were analyzed through the tensile properties. The optimum value of tensile strength and modulus was achieved when 2.0 wt % of CNCs content was incorporated into the matrix. As a conclusion, CNCs can be produced from TLWF and can be used as a filler for PLA at the 2.0 wt % of CNCs loading.

Abstrak tesis yang dikemukakan kepada Senat Universiti Putra Malaysia
sebagai memenuhi keperluan untuk ijazah Master Sains

PENGEKSTRAKAN DAN PENCIRIAN NANOKRISTAL SELULOSA DARI SERAT SISA DAUN TEH SEBAGAI PENGISI DALAM POLI(ASID LAKTIK) BIOKOMPOSIT NANO

Oleh

NUR HAYATI BINTI ABDUL RAHMAN

September 2018

Pengerusi: Nor Azowa binti Ibrahim, PhD
Fakulti: Sains

Poli(asid laktik) (PLA) dikenali sebagai bahan yang sangat berguna dalam menggantikan polimer berasaskan petrokimia dalam sektor pembungkusan disebabkan oleh keterbiodegradan dan ciri mekanikal yang baik. Walaupun ciri PLA yang mengagumkan, kelenturan yang rendah itu sendiri telah mengehadkan penggunaan bahan ini. Oleh itu, nanokristal selulosa (CNCs) diekstrak daripada serat sisa daun teh (TLWF) untuk dimasukkan ke dalam PLA, meningkatkan prestasi nanokomposit polimer sambil mengekalkan alam sekitar dengan selamat.

Kajian ini dijalankan untuk meneliti penggunaan TLWF sebagai sumber penghasilan CNCs dan digunakan sebagai pengisi pada PLA. TLWF pertama kali dirawat dengan alkali, diikuti dengan peluntur sebelum dihidrolisis dengan asid sulfurik pekat. Hasil yang diperolehi selepas setiap langkah rawatan kimia dicirikan. Dari pengubah spektroskopi inframerah Fourier (FTIR), puncak pada 1716 cm^{-1} mewakili regangan C=O hilang dalam spektrum selepas rawatan alkali dan peluntur menunjukkan bahawa hemiselulosa dan lignin hampir dibuang dari serat. Pengurangan intensiti pada 1236 cm^{-1} yang mewakili kepada getaran regangan C-O pada lignin dan xylan berlaku disebabkan penurunan kandungan lignin dan sedikit hemiselulosa dari TLWF. Sementara itu, kestabilan terma CNCs berkurangan disebabkan penggantian kumpulan hidroksil oleh kumpulan sulfat semasa hidrolisis. Mikroskop imbasan electron (SEM) menunjukkan defibrilasi gentian berlaku selepas rawatan, meningkatkan kecenderungan serat untuk hidrolisis asid. Indeks penghabluran serat juga meningkat dari 1.5 % hingga 83.1 % dan dari mikroskop transmisi elektron (TEM), CNCs berbentuk seperti rod dengan diameter purata 7.97 ± 1.09 nm ditunjukkan.

Kemudian, CNCs yang terhasil digunakan sebagai pengisi dalam biokomposit nano PLA melalui kaedah tuangan pelarut. Kandungan CNC yang berbeza antara 0.5 hingga 5 % berat digunakan. Sifat mekanikal filem dianalisis melalui sifat tegangan. Nilai optimum kekuatan tegangan dan modulus dicapai apabila 2.0 wt % kandungan CNC dimasukkan ke dalam matrik. Sebagai kesimpulan, CNCs boleh dihasilkan dari TLWF dan boleh digunakan sebagai pengisi untuk PLA pada jumlah 2.0 wt % CNCs.

ACKNOWLEDGEMENTS

In the name of Allah, The Most Gracious, The Most Merciful.

Firstly, all praise and thanks to Allah because giving me the strength and the opportunity to complete my master project successfully. I would like to express my sincere gratitude to my supervisor Assoc. Prof. Dr. Nor Azowa binti Ibrahim for her valuable guidance along my Master studies. Besides that, I would like to acknowledge to my co-supervisor Dr. Norizah binti Abdul Rahman for her encouragement whenever I had problems to finish this research.

I also thank Dr. Chieng Buong Woei who guides me in an experiment and paper writing until the paper with the title "Extraction and Characterization of Cellulose Nanocrystals from Tea Leaf Waste Fibers" was successfully published on November 2017. My extended gratitude goes to all my labmates who are willing to share their knowledge and experience. In addition, I thank all the staff members at Chemistry Department and Institute Bioscience for their commitment to run and teach me how to analyze the samples.

My sincere appreciation is then to the Ministry of Education, Malaysia, for the MyMaster scholarship, under MYBRAIN15. Deepest gratitude goes to my beloved family for their encouragement and understanding throughout my study at UPM. Lastly, thanks to all my housemates (Casa Impianians) and friends who always support me during this special journey in my life. Thank you.

I certify that a Thesis Examination Committee has met on 26 September 2018 to conduct the final examination of Nur Hayati binti Abdul Rahman on her thesis entitled "Extraction and Characterization of Cellulose Nanocrystals from Tea Leaf Waste Fibers as A Filler in Poly(Lactic Acid) Bio-Nanocomposites" in accordance with the Universities and University Colleges Act 1971 and the Constitution of the Universiti Putra Malaysia [P.U.(A) 106] 15 March 1998. The Committee recommends that the student be awarded the Master of Science.

Members of the Thesis Examination Committee were as follows:

Abdul Halim bin Abdullah, PhD
Associate Professor
Faculty of Science
Universiti Putra Malaysia
(Chairman)

Mansor bin Hj Ahmad @ Ayob, PhD
Professor Dr.
Faculty of Science
Universiti Putra Malaysia
(Internal Examiner)

Ishak bin Ahmad, PhD
Professor Dr.
Universiti Kebangsaan Malaysia
(External Examiner)

RUSLI HAJI ABDULLAH, PhD
Professor and Deputy Dean
School of Graduate Studies
Universiti Putra Malaysia

Date: 31 October 2018

This thesis was submitted to the Senate of Universiti Putra Malaysia and has been accepted as fulfillment of the requirement for the degree of Master of Science. The members of the Supervisory Committee were as follows:

Nor Azowa binti Ibrahim, PhD
Associate Professor
Faculty of Science
Universiti Putra Malaysia
(Chairman)

Norizah binti Abdul Rahman, PhD
Senior Lecturer
Faculty of Science
Universiti Putra Malaysia
(Member)

ROBIAH BINTI YUNUS, PhD
Professor and Dean
School of Graduate Studies
Universiti Putra Malaysia

Date:

Declaration by graduate student

I hereby confirm that:

- this thesis is my original work;
- quotations, illustrations and citations have been duly referenced;
- this thesis has not been submitted previously or concurrently for any other degree at any other institutions;
- intellectual property from the thesis and copyright of thesis are fully-owned by Universiti Putra Malaysia, as according to the Universiti Putra Malaysia (Research) Rules 2012;
- written permission must be obtained from supervisor and the office of Deputy Vice-Chancellor (Research and Innovation) before thesis is published (in the form of written, printed or in electronic form) including books, journals, modules, proceedings, popular writings, seminar papers, manuscripts, posters, reports, lecture notes, learning modules or any other materials as stated in the Universiti Putra Malaysia (Research) Rules 2012;
- there is no plagiarism or data falsification/fabrication in the thesis, and scholarly integrity is upheld as according to the Universiti Putra Malaysia (Graduate Studies) Rules 2003 (Revision 2012-2013) and the Universiti Putra Malaysia (Research) Rules 2012. The thesis has undergone plagiarism detection software.

Signature: ______________________ Date: __________________

Name and Matric No.: Nur Hayati binti Abdul Rahman, GS46028

Declaration by Members of Supervisory Committee

This is to confirm that:

- the research conducted and the writing of this thesis was under our supervision;
- supervision responsibilities as stated in the Universiti Putra Malaysia (Graduate Studies) Rules 2003 (Revision 2012-2013) are adhered to.

Signature: ______________________
Name of Chairman of Supervisory Committee: Nor Azowa binti Ibrahim

Signature: ______________________
Name of Member of Supervisory Committee: Norizah binti Abdul Rahman

TABLE OF CONTENTS

LIST OF TABLES

LIST OF FIGURES

LIST OF ABBREVIATIONS

AFM	Atomic Force Microscopy
CNCs	Cellulose nanocrystals
CNFs	Cellulose nanofibrils
CNWs	Cellulose nanowhiskers
DTG	Derivative Thermogravimetric
E-SEM	Emission-Scanning Electron Microscopy
FAO	Food and Agricultural Organization
FDA	Food and Drug Administration
FTIR	Fourier Transform Infrared
MCC	Microcrystalline cellulose
NCC	Nanocrystalline cellulose
PCL	Polycaprolactone
PE	Polyethylene
PGA	Poly(glycolic acid)
PLA	Poly(lactic acid)
PP	Polypropylene
PVA	Poly(vinyl alcohol)
SEM	Scanning Electron Microscopy
TEM	Transmission Electron Microscopy
T_g	Glass transition temperature
TGA	Thermogravimetric Analysis
TLWF	Tea leaf waste fibers
T_m	Melting temperature
UV	Ultraviolet
XRD	X-ray Diffraction

CHAPTER 1

INTRODUCTION

1.1 General Background

Most of the petroleum-based polymers were used in packaging sectors to conserve the food from degradation process by physical or chemical impurities, microbial contamination, and loss of scent while maintaining the quality of material during its extended shelf life. However, these petroleum-based polymers are non-renewable and non-compostable which can cause a serious environmental and disposal problem (Talegaonkar et al., 2017).

Due to great concerns about this issues, recycle strategy had introduced to the community but normally plastic packaging products cannot always be recycled. A lot of plastic-based packaging materials are disposed of in landfills or combusted into the environment which can emit noxious greenhouse gases because they basically consist of polyethylene and need up to 1,000 years to decay (Malathi et al.,2014).

The production of new products needs to move to sustainable materials basis which is more compliant with the environment and less dependence on fossil fuels. One of the products that fit well with all the characteristics is bio-based polymer composites because they can moderate the issues of sustainability, contributing the possibility of renewability, biodegradation, environmentally benign and a step away from hazardous substances. This is intentional to mitigate the anthropogenic activities from making a continuous destruction of the earth.

In the 1990s, a lot of new biodegradable materials had introduced to the community such as polyurethane and solvents from soy oil sources; adhesives from soy protein; lubricants based on vegetable oil; organic acids based on crop materials; and biocomposites processing from natural cellulose fibers mixed with petrochemical-based polymers such as polyethylene (PE) and polypropylene (PP) or bio-based polymers like PLA, bioresins made from vegetable oils and cellulose esters (Drumright et al., 2000). Among the many kinds of bio-based polymers, PLA is one of the most favorable materials. The biodegradability, biocompatibility, abundant renewable source, transparency, UV stability, excellent mechanical properties, safety, and excellent processability are several unique properties of PLA. Furthermore, the mechanical features such as tensile strength and Young's modulus remark PLA as one of the most encouraging alternatives for petrochemical-based polymers (Frone et al., 2013). However, PLA has its own shortcomings such as brittleness in sufficient strength, low viscosity, medium gas barrier, high moisture sensitivity, and dimensional stability make it not appropriate for load-bearing implementation. Hence, some considerable attempts have been made to upgrade the performance of PLA by initiated the polymer composites with natural fibers. Natural fibers are environmentally friendly, nearly inexhaustible, totally biodegradable, abundantly available, and inexpensive sources. Meanwhile, the hierarchical and multi-level alignment of natural fibers permits the development of nanosize properties (Azizi Samir et al., 2005).

The improvement of bio-nanocomposites performance that entirely degrade in the environment without producing any toxic materials represents the main point of this research. Cellulose nanocrystals have drawn significant attention amongst researchers as promising nanofiller in several polymers. Various terminologies have been stated in the previous works to define these cellulose nanocrystals (CNCs) including nanowhiskers, nanocrystalline cellulose (NCC), monocrystals, microcrystals or microcrystallites (Siqueira et al.,2011).

A variety of agricultural wastes have been studied as natural sources in the CNCs preparation. Even though more variations of natural fibers were examined in detail, the utilization of tea leaves waste fibers (TLWF) as a plant source for the extraction of CNCs has not been reported yet. In tea beverage industries, tea leaves are widely used. However, the usage of tea residue after the process is limited. Thus, one of the suitable solutions to enhance the utilization of TLWF is the extraction of CNCs from the fibers.

Polymer bio-nanocomposites are designated as materials made from bio-based polymers constituting reinforcing agent with at least one of the dimensions is smaller than 100 nm and combined together by using some techniques (Oksmann et al., 2006). Generally, the bio-based polymer will play as a matrix and nanoscale inorganic or organic filler as reinforcement. The role of the matrix is to surround and guide the reinforcements by protecting their corresponding positions while the reinforcements transmit the special physical features to strengthen the matrix behaviors. The use of nanofiller as a reinforcement to these bio-based polymers may advance not only to the performances of the products but also open new capabilities in nanotechnology industries.

1.2 Problem Statement

Nowadays, the implementation of bio-based polymer instead of petroleum-based polymer had reduced the environmental pollutions because of its biodegradability characteristic. However, some drawbacks of PLA such as brittleness, low mechanical and hydrophobic properties have limit itself to be used in some applications. These concerns can be attained by using a filler to improve its performance.

Tea leaf waste fiber (TLWF) is a hidden potential of natural fiber that can be extracted to CNCs to be used as a filler on the bio-based polymer. TLWF is a residue product of tea-leave processing, extracted after drying and chopping off the leaves. Although the cellulose content of tea residue is reported low from the previous finding by Tutus et al. (2015), but the negative environmental impacts created by simply disposing of TLWF could be reduced by extracting CNCs from this source. Besides that, there is no report about extraction of CNCs from TLWF.TLWF was also chosen due to their renewable nature, low density, highly compatible, high modulus, readily available and ecological friendliness.

However, the hydrophilicity property of CNCs reduced its compatibility with the PLA (hydrophobic polymer). Thus, the pretreatment of fibers was conducted to overcome this problem. In addition, the controlled sulfuric acid hydrolysis technique was conducted to extract CNCs. The resulting product will be used as a filler in PLA, initiating PLA/CNCs bio-nanocomposites *via* solvent casting method. Lastly, the performance of the films was studied.

Objectives

The objectives of this research are;

1. To extract the CNCs from tea leaf waste fibers *via* controlled sulphuric acid hydrolysis.
2. To characterize the CNCs by chemical composition, FTIR spectroscopy, thermal properties, X-ray Diffraction analysis, Scanning Electron Microscopy and Transmission Electron Microscopy after each process.
3. To prepare the PLA/CNCs bio-nanocomposites and characterized their mechanical, thermal, and morphological properties after CNCs were added.

REFERENCES

Abe, K., Iwamoto, S., & Yano, H. (2007). Obtaining cellulose nanofibers with a uniform width of 15 nm from wood. *Biomacromolecules*, *8*(10), 3276–3278.

Ahvazi, B., & Ngo, T. D. (2018). Application of Lignins in Formulation and Manufacturing Bio-Based Polyurethanes by 31P NMR Spectroscopy. *In Lignin-Trends and Applications. InTech.*

Akhlaghi, S. P., Tiong, D., Berry, R. M., & Tam, K. C. (2014). Comparative release studies of two cationic model drugs from different cellulose nanocrystal derivatives. *European Journal of Pharmaceutics and Biopharmaceutics*, *88*(1), 207–215.

Alemdar, A., & Sain, M. (2008). Isolation and characterization of nanofibers from agricultural residues - Wheat straw and soy hulls. *Bioresource Technology*, *99*(6), 1664–1671.

Antal, M. J., Varhegyi, G., & Jakab, E. (1998). Cellulose pyrolysis kinetics: revisited. *Industrial & Engineering Chemistry Research*, *37*(4), 1267–1275.

Araki, J., Wada, M., Kuga, S., & Okano, T. (1998). Flow properties of microcrystalline cellulose suspension prepared by acid treatment of native cellulose. *Colloids and Surfaces A: Physicochemical and Engineering Aspects*, *142*(1), 75–82.

Araki, J., Wada, M., Kuga, S., & Okano, T. (2000). Birefringent glassy phase of a cellulose microcrystal suspension. *Langmuir*, *16*(6), 2413–2415.

Azizi Samir, M. A. S., Alloin, F., Gorecki, W., Sanchez, J. Y., & Dufresne, A. (2004). Nanocomposite polymer electrolytes based on poly (oxyethylene) and cellulose nanocrystals. *The Journal of Physical Chemistry B*, *108*(30), 10845–10852.

Azizi Samir, M. A. S., Alloin, F., & Dufresne, A. (2005). Review of recent research into cellulosic whiskers, their properties and their application in nanocomposite field. *Biomacromolecules*, *6*(2), 612–626.

Bajpai, P. K., Singh, I., & Madaan, J. (2014). Development and characterization of PLA-based green composites: A review. *Journal of Thermoplastic Composite Materials*, *27*(1), 52–81.

Beck-candanedo, S., Roman, M., & Gray, D. G. (2005). Effect of reaction conditions on the properties and behavior of wood cellulose nanocrystal suspensions. *Biomacromolecules*, 6(2), 1048-1054.

Benavides, E. E. U. (2011). Cellulose nanocrystals properties and applications in renewable nanocomposites. *(Doctoral Dissertation, Clemson University).*

Bendahou, A., Habibi, Y., Kaddami, H., & Dufresne, A. (2009). Physico-chemical characterization of palm from phoenixdactylifera–L, preparation of cellulose whiskers and natural rubber–based nanocomposites. *Journal of Biobased Materials and Bioenergy*, *3*(1), 81–90.

Bettaieb, F., Khiari, R., Dufresne, A., Mhenni, M. F., & Belgacem, M. N. (2015). Mechanical and thermal properties of Posidonia oceanica cellulose nanocrystal reinforced polymer. *Carbohydrate Polymers*, *123*, 99–104.

Birnin-Yauri, A. U., Ibrahim, N. A., Zainuddin, N., Abdan, K., Then, Y. Y., & Chieng, B. W. (2016). Influence of kenaf core fiber incorporation on the mechanical performance and dimensional stability of oil palm fiber reinforced poly (lactic acid) hybrid biocomposites. *BioResources*, *11*(2), 3332-3355.

Bismarck, A., Aranberri-Askargorta, I. Springer, J., Lampke, T., Wielage, B., Stamboulis, A., Shenderovich, I., & Limbach, H. H. (2002). Surface characterization of flax, hemp and cellulose fibers; surface properties and the water uptake behavior. *Polymer Composites*, *23*(5), 872–894.

Bledzki, A. K., Reihmane, S., & Gassan, J. (1996). Properties and modification methods for vegetable fibers for natural fiber composites. *Journal of Applied Polymer Science*, *59*(8), 1329–1336.

Bledzki, A. K., & Gassan, J. (1999). Composites reinforced with cellulose based fibres. *Progress in Polymer Science*, *24*(2), 221–274.

Bledzki, A. K., Mamun, A. A., & Volk, J. (2010). Physical, chemical and surface properties of wheat husk, rye husk and soft wood and their polypropylene composites. *Composites Part A: Applied Science and Manufacturing*, *41*(4), 480–488.

Börjesson, M., & Westman, G. (2015). Crystalline nanocellulose preparation, modification, and properties. In *Cellulose-fundamental aspects and current trends*. InTech.

Brinchi, L., Cotana, F., Fortunati, E., & Kenny, J. M. (2013). Production of nanocrystalline cellulose from lignocellulosic biomass: Technology and applications. *Carbohydrate Polymers*, *94*(1), 154–169.

Cao, S. L., Xu, H., Li, X. H., Lou, W. Y., & Zong, M. H. (2015). Papain@ magnetic nanocrystalline cellulose nanobiocatalyst: a highly efficient biocatalyst for dipeptide biosynthesis in deep eutectic solvents. *ACS Sustainable Chemistry & Engineering*, *3*(7), 1589–1599.

Cao, X., Dong, H., & Li, C. M. (2007). New nanocomposite materials reinforced with flax cellulose nanocrystals in waterborne polyurethane. *Biomacromolecules*, *8*(3), 899–904.

Cao, X., Chen, Y., Chang, P. R., Stumborg, M., & Huneault, M. A. (2008). Green composites reinforced with hemp nanocrystals in plasticized starch. *Journal of Applied Polymer Science*, *109*(6), 3804–3810.

Chan, C. H., Chia, C. H., Zakaria, S., Ahmad, I., Dufresne, A., & Tshai, K. Y. (2015). Low filler content cellulose nanocrystal and graphene oxide reinforced polylactic acid film composites. *Polymers Research Journal*, *9*(1), 165.

Chang, K. (2015). World tea production and trade current and future development. *Food and Agricultural Organization of the United Nations, Rome.*

Cheng, M., Qin, Z., Liu, Y., Qin, Y., Li, T., Chen, L., & Zhu, M. (2014). Efficient extraction of carboxylated spherical cellulose nanocrystals with narrow distribution through hydrolysis of lyocell fibers by using ammonium persulfate as an oxidant. *J. Mater. Chem. A*, *2*(1), 251–258.

Chieng, B., Lee, S., Ibrahim, N., Then, Y., & Loo, Y. (2017). Isolation and Characterization of Cellulose Nanocrystals from Oil Palm Mesocarp Fiber. *Polymers*, *9*(8), 355.

Cohn, D., & Hotovely Salomon, A. (2005). Designing biodegradable multiblock PCL/PLA thermoplastic elastomers. *Biomaterials*, *26*(15), 2297–2305.

Conn, R. E., Kolstad, J. J., Borzelleca, J. F., Dixler, D. S., Filer Jr, L. J., LaDu Jr, B. N., & Pariza, M. W. (1995). Safety assessment of polylactide (PLA) for use as a food-contact polymer. *Food and Chemical Toxicology*, *33*(4), 273–283.

Costa, L. A. D. S., Fonsêca, A. F., Pereira, F. V., & Druzian, J. I. (2015). Extraction and Characterization of Cellulose Nanocrystals from Stover. *Cellulose Chemistry and Technology*, *49*(2), 127–133.

Darder, M., Aranda, P., & Ruiz-Hitzky, E. (2007). Bionanocomposites: a new concept of ecological, bioinspired, and functional hybrid materials. *Advanced Materials*,*19*(10), 1309–1319.

De Menezes, A. J., Siqueira, G., Curvelo, A. A., & Dufresne, A. (2009). Extrusion and characterization of functionalized cellulose whiskers reinforced polyethylene nanocomposites. *Polymer*, *50*(19), 4552–4563.

de Oliveira, F. B., Bras, J., Pimenta, M. T. B., da Silva Curvelo, A. A., & Belgacem, M. N. (2016). Production of cellulose nanocrystals from sugarcane bagasse fibers and pith. *Industrial Crops and Products*, *93*, 48–57.

Dizadji, N., & Anaraki, N. A. (2011). Adsorption of chromium and copper in aqueous solutions using tea residue. *International Journal of Environmental Science & Technology*, *8*(3), 631–638.

Domingues, R. M., Gomes, M. E., & Reis, R. L. (2014). The potential of cellulose nanocrystals in tissue engineering strategies. *Biomacromolecules*, *15*(7), 2327–2346.

Dong, H., Strawhecker, K. E., Snyder, J. F., Orlicki, J. A., Reiner, R. S., & Rudie, A. W. (2012). Cellulose nanocrystals as a reinforcing material for electrospun poly (methyl methacrylate) fibers: Formation, properties and nanomechanical characterization. *Carbohydrate Polymers*, *87*(4), 2488–2495.

Dong, S., & Roman, M. (2007). Fluorescently labeled cellulose nanocrystals for bioimaging applications. *Journal of the American Chemical Society*, *129*(45), 13810–13811.

Drumright, R. E., Gruber, P. R., & Henton, D. E. (2000). Polylactic acid technology. *Advanced Materials*, *12*(23), 1841–1846.

El Achaby, M., Kassab, Z., Aboulkas, A., Gaillard, C., & Barakat, A. (2018). Reuse of red algae waste for the production of cellulose nanocrystals and its application in polymer nanocomposites. *International Journal of Biological Macromolecules*, *106*, 681–691.

Elazzouzi-Hafraoui, S., Nishiyama, Y., Putaux, J. L., Heux, L., Dubreuil, F., & Rochas, C. (2007). The shape and size distribution of crystalline nanoparticles prepared by acid hydrolysis of native cellulose. *Biomacromolecules*, *9*(1), 57-65.

Ersoy, S., & Küçük, H. (2009). Investigation of industrial tea-leaf-fibre waste material for its sound absorption properties. *Applied Acoustics*, *70*(1), 215–220.

Favier, V., Chanzy, H., & Cavaille, J. Y. (1995). Polymer nanocomposites reinforced by cellulose whiskers. *Macromolecules*,*28*(18), 6365–6367.

Filson, P. B., & Dawson-Andoh, B. E. (2009). Sono-chemical preparation of cellulose nanocrystals from lignocellulose derived materials. *Bioresource Technology*, *100*(7), 2259–2264.

Fiore, V., Scalici, T., Nicoletti, F., Vitale, G., Prestipino, M., & Valenza, A. (2016). A new eco-friendly chemical treatment of natural fibres: Effect of sodium bicarbonate on properties of sisal fibre and its epoxy composites. *Composites Part B: Engineering*, *85*, 150–160.

Fisher, T., Hajaligol, M., Waymack, B., & Kellogg, D. (2002). Pyrolysis behavior and kinetics of biomass derived materials. *Journal of analytical and applied pyrolysis*, *62*(2), 331-349.

Fortunati, E., Armentano, I., Zhou, Q., Iannoni, A., Saino, E., Visai, L., Berglund. L.A., & Kenny,J.M.(2012). Multifunctional bionanocomposite films of poly (lactic acid), cellulose nanocrystals and silver nanoparticles. *Carbohydrate Polymers*, *87*(2), 1596-1605.

Frone, A. N., Berlioz, S., Chailan, J.-F., & Panaitescu, D. M. (2013). Morphology and thermal properties of PLA - cellulose nanofibers composites. *Carbohydrate Polymers*, 91(1), 377-384.

Garcia de Rodriguez, N. L., Thielemans, W., & Dufresne, A. (2006). Sisal cellulose whiskers reinforced polyvinyl acetate nanocomposites. *Cellulose*, *13*(3), 261-270.

Gassan, J., & Bledzki, A. K. (1997). The influence of fiber-surface treatment on the mechanical properties of jute-polypropylene composites. *Composites Part A: Applied Science and Manufacturing*, *28*(12), 1001–1005.

Gazzotti, S., Farina, H., Lesma, G., Rampazzo, R., Piergiovanni, L., Ortenzi, M. A., & Silvani, A. (2017). Polylactide/cellulose nanocrystals: The in situ polymerization approach to improved nanocomposites. *European Polymer Journal*, *94*, 173–184.

Grishkewich, N., Mohammed, N., Tang, J., & Tam, K. C. (2017). Recent advances in the application of cellulose nanocrystals. *Current Opinion in Colloid & Interface Science*, *29*, 32-45.

Grunert, M., & Winter, W. T. (2002). Nanocomposites of cellulose acetate butyrate reinforced with cellulose nanocrystals. *Journal of Polymers and the Environment*, *10*(1-2), 27-30.

Haafiz, M. M., Hassan, A., Khalil, H. A., Khan, I., Inuwa, I. M., Islam, M. S., Hossain, M.S., Syakir, M.I., &Fazita, M.R.N. (2015). Bionanocomposite based on cellulose nanowhisker from oil palm biomass-filled poly (lactic acid). *Polymer Testing*, *48*, 133–139.

Habibi, Y., Goffin, A. L., Schiltz, N., Duquesne, E., Dubois, P., & Dufresne, A. (2008). Bionanocomposites based on poly (ε-caprolactone)-grafted cellulose nanocrystals by ring-opening polymerization. *Journal of Materials Chemistry*, *18*(41), 5002-5010.

Habibi, Y., Lucia, L. A., & Rojas, O. J. (2010). Cellulose nanocrystals: chemistry, self-assembly, and applications. *Chemical reviews*, *110*(6), 3479-3500.

He, X., Male, K. B., Nesterenko, P. N., Brabazon, D., Paull, B., & Luong, J. H. (2013). Adsorption and desorption of methylene blue on porous carbon monoliths and nanocrystalline cellulose. *ACS Applied Materials & Interfaces*, *5*(17), 8796–8804.

Hosseini, S. E., & Abdul Wahid, M. (2014). The role of renewable and sustainable energy in the energy mix of Malaysia. *International Journal of Energy Research*, *38*(14), 1769–1792.

Iwamoto, S., Kai, W., Isogai, A., & Iwata, T. (2009). Elastic modulus of single cellulose microfibrils from tunicate measured by atomic force microscopy. *Biomacromolecules*, *10*(9), 2571–2576.

Jamshidian, M., Tehrany, E. A., Imran, M., Jacquot, M., & Desobry, S. (2010). Poly-Lactic Acid: Production, applications, nanocomposites, and release studies. *Comprehensive Reviews in Food Science and Food Safety*, *9*(5), 552–571.

Johar, N., Ahmad, I., & Dufresne, A. (2012). Extraction, preparation and characterization of cellulose fibres and nanocrystals from rice husk. *Industrial Crops and Products*, *37*(1), 93–99.

Jonoobi, M., Harun, J., Mathew, A. P., Hussein, M. Z. B., &Oksman, K. (2010). Preparation of cellulose nanofibers with hydrophobic surface characteristics. Cellulose, 17(2), 299 -307.

Jonoobi, M., Oladi, R., Davoudpour, Y., Oksman, K., Dufresne, A., Hamzeh, Y., & Davoodi, R. (2015). Different preparation methods and properties of nanostructured cellulose from various natural resources and residues: a review. *Cellulose*, *22*(2), 935–969.

Kalia, S., Kaith, B. S., & Kaur, I. (2009). Pretreatments of natural fibers and their application as reinforcing material in polymer composites-a review. *Polymer Engineering and Science*, *49(7)*, 1253-1272

Kargarzadeh, H., Ahmad, I., Abdullah, I., Dufresne, A., Zainudin, S. Y., & Sheltami, R. M. (2012). Effects of hydrolysis conditions on the morphology, crystallinity, and thermal stability of cellulose nanocrystals extracted from kenaf bast fibers. *Cellulose*, *19*(3), 855–866.

Khalil, H. A., Bhat, I. U. H., Jawaid, M., Zaidon, A., Hermawan, D., & Hadi, Y. S. (2012). Bamboo fibre reinforced biocomposites: A review. *Materials & Design*, *42*, 353-368.

Khalil, H. A., Davoudpour, Y., Islam, M. N., Mustapha, A., Sudesh, K., Dungani, R., & Jawaid, M. (2014). Production and modification of nanofibrillated cellulose using various mechanical processes: a review. *Carbohydrate polymers*, *99*, 649-665.

Khoo, R. Z., Ismail, H., & Chow, W. S. (2016). Thermal and morphological properties of poly (lactic acid)/nanocellulose nanocomposites. *Procedia Chemistry*, *19*, 788–794.

Klemm, D., Schumann, D., Kramer, F., Heßler, N., Hornung, M., Schmauder, H. P., & Marsch, S. (2006). Nanocelluloses as innovative polymers in research and application. *In Polysaccharides Ii*, 49–96.

Koegler, W. S., & Griffith, L. G. (2004). Osteoblast response to PLGA tissue engineering scaffolds with PEO modified surface chemistries and demonstration of patterned cell response. *Biomaterials*, *25*(14), 2819-2830.

Ku, H., Wang, H., Pattarachaiyakoop, N., & Trada, M. (2011). A review on the tensile properties of natural fiber reinforced polymer composites. *Composites Part B: Engineering*, *42*(4), 856–873.

Kumar, A., Negi, Y. S., Choudhary, V., & Bhardwaj, N. K. (2014). Characterization of cellulose nanocrystals produced by acid-hydrolysis from sugarcane bagasse as agro-waste. *Journal of Materials Physics and Chemistry*, *2*(1), 1–8.

Kuorwel, K. K., Cran, M. J., Sonneveld, K., Miltz, J., & Bigger, S. W. (2011). Antimicrobial activity of biodegradable polysaccharide and protein-based films containing active agents. *Journal of Food Science*, *76*(3), 90-102.

Lamaming, J., Hashim, R., Sulaiman, O., Leh, C. P., Sugimoto, T., & Nordin, N. A. (2015). Cellulose nanocrystals isolated from oil palm trunk. *Carbohydrate Polymers*, *127*, 202–208.

Le Troëdec, M., Sedan, D., Peyratout, C., Bonnet, J. P., Smith, A., Guinebretiere, R., Gloaguen, V., & Krausz, P. (2008). Influence of various chemical treatments on the composition and structure of hemp fibers. *Composites Part A*:*Applied Science and Manufacturing*, *39(3)*, 514-522.

Li, J., Wei, X., Wang, Q., Chen, J., Chang, G., Kong, L., Su, J., & Liu, Y. (2012). Homogeneous isolation of nanocellulose from sugarcane bagasse by high pressure homogenization. *Carbohydrate Polymers*, *90*(4), 1609–1613.

Liew, S. Y., Walsh, D. A., & Thielemans, W. (2013). High total-electrode and mass-specific capacitance cellulose nanocrystal-polypyrrole nanocomposites for supercapacitors. *Rsc Advances*, *3*(24), 9158-9162.

Lin, N., Huang, J., Chang, P. R., Feng, J., & Yu, J. (2011). Surface acetylation of cellulose nanocrystal and its reinforcing function in poly (lactic acid). *Carbohydrate Polymers*, *83*(4), 1834–1842.

Lin, N., & Dufresne, A. (2014). Nanocellulose in biomedicine: Current status and future prospect. *European Polymer Journal*, *59*, 302–325.

Liu, D. Y., Yuan, X. W., Bhattacharyya, D., & Easteal, A. J. (2010). Characterisation of solution cast cellulose nanofibre-reinforced poly (lactic acid). *Express Polymer Letters*, *4*(1), 26-31.

Liu, M., Zhang, Y., & Zhou, C. (2013). Nanocomposites of halloysite and polylactide. *Applied Clay Science*, *75*, 52-59.

Liu, S., & Tang, Z. (2010). Nanoparticle assemblies for biological and chemical sensing. *Journal of Materials Chemistry*, *20*(1), 24–35.

Liu, Z., & Huang, H. (2016). Preparation and characterization of cellulose composite hydrogels from tea residue and carbohydrate additives. *Carbohydrate Polymers*, *147*, 226–233.

Lu, P., & Hsieh, Y. L. (2010). Preparation and properties of cellulose nanocrystals: rods, spheres, and network. *Carbohydrate polymers*, *82*(2), 329-336.

Lu, Y., Weng, L., & Cao, X. (2005). Biocomposites of plasticized starch reinforced with cellulose crystallites from cottonseed linter. *Macromolecular Bioscience*, *5*(11), 1101–1107.

Lunt, J. (1998). Large-scale production, properties and commercial applications of polylactic acid polymers. *Polymer Degradation and Stability*, *59*(1), 145–152.

Malainine, M. E., Mahrouz, M., & Dufresne, A. (2005). Thermoplastic nanocomposites based on cellulose microfibrils from Opuntia ficus-indica parenchyma cell. *Composites Science and Technology*, *65*(10), 1520–1526.

Malathi, A. N., Santhosh, K. S., & Nidoni, U. (2014). Recent trends of biodegradable polymer: biodegradable films for food packaging and application of nanotechnology in biodegradable food packaging. *Current Trends in Technology and Science*, *3*(2), 73-79.

Malkoc, E., & Nuhoglu, Y. (2006). Removal of Ni (II) ions from aqueous solutions using waste of tea factory: Adsorption on a fixed-bed column. *Journal of Hazardous Materials*, *135*(1–3), 328–336.

Man, Z., Muhammad, N., Sarwono, A., Bustam, M. A., Kumar, M. V., & Rafiq, S. (2011). Preparation of Cellulose Nanocrystals Using an Ionic Liquid. *Journal of Polymers and the Environment*, *19*(3), 726–731.

Mandal, A., & Chakrabarty, D. (2011). Isolation of nanocellulose from waste sugarcane bagasse (SCB) and its characterization. *Carbohydrate Polymers*, *86*(3), 1291–1299.

Mason, S., Reinecke, C. J., Kulik, W., Van Cruchten, A., Solomons, R., & van Furth, A.M.T. (2016). Cerebrospinal fluid in tuberculous meningtis exhibits only L-enantiomer of lactic acid. *BMC infectious diseases, 16(1)*, 251.

Mazlita, Y., Lee, H. V., & Hamid, S. B. A. (2016). Preparation of cellulose nanocrystals bio-polymer from agro-industrial wastes: Separation and characterization. *Polymers & Polymer Composites*, *24*(9), 719.

Moon, R. J., Martini, A., Nairn, J., Simonsen, J., & Youngblood, J. (2011). Cellulose nanomaterials review: structure, properties and nanocomposites. *Chemical Society Reviews*,*40*(7), 3941–3994.

Morán, J. I., Alvarez, V. A., Cyras, V. P., & Vázquez, A. (2008). Extraction of cellulose and preparation of nanocellulose from sisal fibers. *Cellulose*, *15*(1), 149–159.

Moriana, R.; Vilaplana, F.; Ek, M. Cellulose nanocrystals from forest residues as reinforcing agents for

composites: A study from macro- to nano-dimensions. Carbohydr. Polym. **2016**, 139, 139–149.

Morin, A., & Dufresne, A. (2002). Nanocomposites of chitin whiskers from Riftia tubes and poly (caprolactone). *Macromolecules*, *35*(6), 2190–2199.

Nadanathangam, V., & Satyamurthy, P. (2011). Preparation of spherical nanocellulose by anaerobic microbial consortium. *In 2 Nd International Conference on Biotechnology and Food Science*, 181–183.

Naduparambath, S., T.V., J., Shaniba, V., M.P., S., Balan, A. K., & Purushothaman, E. (2018). Isolation and characterisation of cellulose nanocrystals from sago seed shells. *Carbohydrate Polymers*, 180, 13-20.

Neto, W. P. F., Silvério, H. A., Dantas, N. O., & Pasquini, D. (2013). Extraction and characterization of cellulose nanocrystals from agro-industrial residue–Soy hulls. *Industrial Crops and Products*, *42*, 480–488.

Nickerson, R. F., & Habrle, J. A. (1947). Cellulose intercrystalline structure. *Industrial & Engineering Chemistry*, *39*(11), 1507–1512.

Oksman, K., Mathew, A. P., Bondeson, D., & Kvien, I. (2006). Manufacturing process of cellulose whiskers/polylactic acid nanocomposites. *Composites Science and Technology*, *66*(15), 2776–2784.

Peresin, M. S., Habibi, Y., Zoppe, J. O., Pawlak, J. J., & Rojas, O. J. (2010). Nanofiber Composites of Polyvinyl Alcohol and Cellulose Nanocrystals: Manufacture and Characterisation. *Biomacromolecules*, *11*(3), 674–681.

Petersson, L., Kvien, I., & Oksman, K. (2007). Structure and thermal properties of poly (lactic acid)/cellulose whiskers nanocomposite materials. *Composites Science and Technology*, *67*(11–12), 2535–2544.

Pielichowski, K., & Njuguna, J. (2005). Thermal degradation of polymeric materials. *iSmithers Rapra Publishing.*

Qian, S., Zhang, H., Yao, W., & Sheng, K. (2018). Effects of bamboo cellulose nanowhisker content on the morphology, crystallization, mechanical, and thermal properties of PLA matrix biocomposites. *Composites Part B: Engineering*, *133*, 203–209.

Qu, P., Gao, Y., Wu, G., & Zhang, L. (2010). Nanocomposites of poly (lactic acid) reinforced with cellulose nanofibrils. *BioResources*, *5*(3), 1811–1823.

Rambabu, N., Panthapulakkal, S., Sain, M., & Dalai, A. K. (2016). Production of nanocellulose fibers from pinecone biomass: Evaluation and optimization of chemical and mechanical treatment conditions on mechanical properties of nanocellulose films. *Industrial Crops and Products*, *83*, 746–754.

Ramires, E. C., & Dufresne, A. (2011). A review of cellulose nanocrystals and nanocomposites. *Technical Association of the Pulp and Paper Industry Journal*, *10*(4), 9–16.

Ranby, B. G. (1949). Aqueous colloidal solutions of cellulose micelles. *Acta Chemica Scandinavica*, *3*(5), 649–650.

Rånby, B. G. (1951). Fibrous macromolecular systems. Cellulose and muscle. The colloidal properties of cellulose micelles. *Discussions of the Faraday Society*, *11*, 158-164.

Rayung, M., Ibrahim, N. A., Zainuddin, N., Saad, W. Z., Razak, N. I. A., & Chieng, B. W. (2014). The effect of fiber bleaching treatment on the properties of poly (lactic acid)/oil palm empty fruit bunch fiber composites. *International Journal of Molecular Sciences*, *15*(8), 14728–14742.

Razak, N. I. A., Ibrahim, N. A., Zainuddin, N., Rayung, M., & Saad, W. Z. (2014). The influence of chemical surface modification of kenaf fiber using hydrogen peroxide on the mechanical properties of biodegradable kenaf fiber/poly(Lactic Acid) composites. *Molecules*, *19*(3), 2957–2968.

Reddy, J. P., & Rhim, J. W. (2014). Characterization of bionanocomposite films prepared with agar and paper-mulberry pulp nanocellulose. *Carbohydrate Polymers*, *110*, 480-488.

Rezwan, K., Chen, Q. Z., Blaker, J. J., & Boccaccini, A. R. (2006). Biodegradable and bioactive porous polymer/inorganic composite scaffolds for bone tissue engineering. *Biomaterials*, *27*(18), 3413–3431.

Rhim, J.W., Park, H.M., & Ha, C.S. (2013). Bio-nanocomposites for food packaging applications. *Progress in Polymer Science*, *38*(10–11), 1629–1652.

Rhim, J. W., Reddy, J. P., & Luo, X. (2015). Isolation of cellulose nanocrystals from onion skin and their utilization for the preparation of agar-based bio-nanocomposites films. *Cellulose*, *22*(1), 407–420.

Roohani, M., Habibi, Y., Belgacem, N. M., Ebrahim, G., Karimi, A. N., & Dufresne, A. (2008). Cellulose whiskers reinforced polyvinyl alcohol copolymers nanocomposites. *European polymer journal*, *44*(8), 2489-2498.

Rong, M. Z., Zhang, M. Q., Liu, Y., Yang, G. C., & Zeng, H. M. (2001). The effect of fiber treatment on the mechanical properties of unidirectional sisal-reinforced epoxy composites. *Composites Science and technology*, *61*(10), 1437-1447.

Roman, M., & Winter, W. T. (2004). Effect of sulfate groups from sulfuric acid hydrolysis on the thermal degradation behavior of bacterial cellulose. *Biomacromolecules*, *5*(5), 1671–1677.

Rosli, N. A., Ahmad, I., & Abdullah, I. (2013). Isolation and characterization of cellulose nanocrystals from agave angustifoliafibre. *BioResources*, *8*(2), 1893–1908.

Sanchez-Garcia, M. D., & Lagaron, J. M. (2010). On the use of plant cellulose nanowhiskers to enhance the barrier properties of polylactic acid. *Cellulose*, *17*(5), 987-1004.

Segal, L., Creely, J. J., Martin, A. E., & Conrad, C. M. (1959). An Empirical Method for Estimating the Degree of Crystallinity of Native Cellulose Using the X-Ray Diffractometer. *Textile Research Journal*, *29*(10), 786–794.

Sheltami, R. M., Abdullah, I., Ahmad, I., Dufresne, A., & Kargarzadeh, H. (2012). Extraction of cellulose nanocrystals from mengkuang leaves (Pandanus tectorius). *Carbohydrate Polymers*, *88*(2), 772–779.

Silvério, H. A., Neto, W. P. F., Dantas, N. O., & Pasquini, D. (2013). Extraction and characterization of cellulose nanocrystals from corncob for application as reinforcing agent in nanocomposites. *Industrial Crops and Products*, *44*, 427–436.

Siqueira, G., Abdillahi, H., Bras, J., & Dufresne, A. (2010). High reinforcing capability cellulose nanocrystals extracted from Syngonanthus nitens (Capim Dourado). *Cellulose*, *17*(2), 289-298.

Siqueira, G., Bras, J., & Dufresne, A. (2010). Cellulosic bionanocomposites: A review of preparation, properties and applications. *Polymers*, *2*(4), 728–765.

Siqueira, G., Tapin-Lingua, S., Bras, J., da Silva Perez, D., & Dufresne, A. (2011). Mechanical properties of natural rubber nanocomposites reinforced with cellulosic nanoparticles obtained from combined mechanical shearing, and enzymatic and acid hydrolysis of sisal fibers. *Cellulose*, *18*(1), 57–65.

Sung, S. H., Chang, Y., & Han, J. (2017). Development of polylactic acid nanocomposite films reinforced with cellulose nanocrystals derived from coffee silverskin. *Carbohydrate Polymers*, *169*, 495–503.

Takagi, H., & Asano, A. (2008). Effects of processing conditions on flexural properties of cellulose nanofiber reinforced "' green '" composites. *Composites Part A: Applied Science and Manufacturing*, *39(4)*, 685-689.

Talegaonkar, S., Sharma, H., Pandey, S., Mishra, P. K., & Wimmer, R. (2017). Bionanocomposites: smart biodegradable packaging material for food preservation. In *Food packaging*, 79-110.

Tang, X.,& Alavi, S. (2011). Recent advances in starch, polyvinyl alcohol based polymer blends, nanocomposites and their biodegradability. *Carbohydrate Polymers*, *85*(1), 7–16.

Tappi, T. (2002). 222 om-02: Acid-insoluble lignin in wood and pulp. 2002–2003 TAPPI Test Methods.

Then, Y. Y., Ibrahim, N. A., Zainuddin, N., Ariffin, H., Yunus, W. M. Z. W., & Chieng, B. W. (2014). Surface modifications of oil palm mesocarp fiber by superheated steam, alkali, and superheated steam-alkali for biocomposite applications. *BioResources*, *9*(4), 7467-7483.

Then, Y. Y., Ibrahim, N. A., Zainuddin, N., Chieng, B. W., Ariffin, H., & Wan Yunus, W. M. Z. (2015). Influence of alkaline-peroxide treatment of fiber on the mechanical properties of oil palm mesocarp fiber/poly(butylene succinate) biocomposite. *BioResources*, *10*(1), 1730–1746.

Tkalya, E., Ghislandi, M., Thielemans, W., van der Schoot, P., de With, G., & Koning, C. (2013). Cellulose nanowhiskers templating in conductive polymer nanocomposites reduces electrical percolation threshold 5-fold. *ACS Macro Letters*, *2*(2), 157–163.

Trache, D., Donnot, A., Khimeche, K., Benelmir, R., & Brosse, N. (2014). Physico-chemical properties and thermal stability of microcrystalline cellulose isolated from Alfa fibres. *Carbohydrate Polymers*, *104*, 223–230.

Turbak, A. F., Snyder, F. W., & Sandberg, K. R. (1983). Microfibrillated cellulose, a new cellulose product: properties, uses, and commercial potential. *Applied Polymer Science*, *37*, 815.

Tutuş, A., Kazaskeroğlu, Y., & çiçekler, M. (2015). Evaluation of tea wastes in usage pulp and paper production. *BioResources*, *10*(3), 5395–5406.

Wambua, P., Ivens, J., & Verpoest, I. (2003). Natural fibres: Can they replace glass in fibre reinforced plastics? *Composites Science and Technology*, *63*(9), 1259–1264.

Wang, K., Jiang, J. X., Xu, F., & Sun, R. C. (2009). Influence of steaming pressure on steam explosion pretreatment of Lespedeza stalks (Lespedeza crytobotrya): Part 1. Characteristics of degraded cellulose. *Polymer Degradation and Stability*, *94*(9), 1379–1388.

Wang, S., Sun, J., Jia, Y., Yang, L., Wang, N., Xianyu, Y., Chen, W., Li, X., Cha, R., & Jiang, X. (2016). Nanocrystalline cellulose-assisted generation of silver nanoparticles for Nonenzymatic glucose detection and antibacterial agent. *Biomacromolecules*, *17*(7), 2472–2478.

Wong, S., & Shanks, R. (2009). Biocomposites of Natural Fibers and Poly (3-Hydroxybutyrate) and Copolymers: Improved Mechanical Properties Through Compatibilization at the Interface. *Biodegradable Polymer Blends and Composites from Renewable Resources*, 303–347.

Wu, C. S., & Liao, H. T. (2005). A new biodegradable blends prepared from polylactide and hyaluronic acid. *Polymer*, *46*(23), 10017–10026.

Xiao, B., Sun, X., & Sun, R. (2001). Chemical, structural, and thermal characterizations of alkali-soluble lignins and hemicelluloses, and cellulose from maize stems, rye straw, and rice straw. *Polymer degradation and stability*, *74*(2), 307-319.

Yalinkilic, M. K., Imamura, Y., Takahashi, M., Kalaycioglu, H., Nemli, G., Demirci, Z., & Ozdemir, T. (1998). Biological, physical and mechanical properties of particleboard manufactured from waste tea leaves. *International Biodeterioration & Biodegradation*, *41*(1), 75–84.

Yokohara, T., & Yamaguchi, M. (2008). Structure and properties for biomass-based polyester blends of PLA and PBS. *European Polymer Journal*, *44*(3), 677–685.

Zhou, C., & Wu, Q. (2012). Recent development in applications of cellulose nanocrystals for advanced polymer-based nanocomposites by novel fabrication strategies. *In Nanocrystals-Synthesis, Characterization and Applications. Intech.*

Zhou, Y., Fuentes-Hernandez, C., Khan, T. M., Liu, J. C., Hsu, J., Shim, J. W., Dindar, A., Youngblood, J.P., Moon, R.J., & Kippelen, B. (2013). Recyclable organic solar cells on cellulose nanocrystal substrates. *Scientific Reports, 3*, 1536.

BIODATA OF STUDENT

Nur Hayati binti Abdul Rahman was born on 15th August 1991 in Tumpat, Kelantan. She started her primary education at Sekolah Kebangsaan Berangan (1), Tumpat and completed her secondary school at MARA Junior Science College (MJSC) Kota Putra, Besut, Terengganu. Then, she pursued her higher education at Gambang Matriculation College, Pahang. She was received her bachelor degree in Bachelor of Science (Honors) - Chemistry from Universiti Putra Malaysia in November 2014. After that, she continued her study for Master Degree in Materials Science under the supervision of Associate Prof. Dr Nor Azowa binti Ibrahim.

LIST OF PUBLICATION

Abdul Rahman, N. H., Chieng, B. W., Ibrahim, N. A., & Abdul Rahman, N. (2017). Extraction and Characterization of Cellulose Nanocrystals from Tea Leaf Waste Fibers. *Polymers*, *9*(11), 588. doi:10.3390/polym9110588.

UNIVERSITI PUTRA MALAYSIA

STATUS CONFIRMATION FOR THESIS / PROJECT REPORT AND COPYRIGHT

ACADEMIC SESSION : ________________

TITLE OF THESIS / PROJECT REPORT :

____

____

____

NAME OF STUDENT :

____

I acknowledge that the copyright and other intellectual property in the thesis/project report belonged to Universiti Putra Malaysia and I agree to allow this thesis/project report to be placed at the library under the following terms:

1. This thesis/project report is the property of Universiti Putra Malaysia.

2. The library of Universiti Putra Malaysia has the right to make copies for educational purposes only.

3. The library of Universiti Putra Malaysia is allowed to make copies of this thesis for academic exchange.

I declare that this thesis is classified as:

*Please tick (√)

☐	**CONFIDENTIAL**	(Contain confidential information under Official Secret Act 1972).
☐	**RESTRICTED**	(Contains restricted information as specified by the organization/institution where research was done).
☐	**OPEN ACCESS**	I agree that my thesis/project report to be published as hard copy or online open access.

This thesis is submitted for:

☐ **PATENT**   Embargo from ______________ until ______________
(date) (date)

Approved by:

____
(Signature of Student)
New IC No/ Passport No.:

Date :

____
(Signature of Chairman
of Supervisory Committee)
Name:

Date :

[Note : If the thesis is CONFIDENTIAL or RESTRICTED, please attach with the letter from the organization/institution with period and reasons for confidentially or restricted.]