

CHARACTERIZATION AND PROPERTIES OF SODIUM HYDROXIDE/UREA  
REGENERATED CELLULOSE REINFORCED POLY(3-HYDROXYBUTYRATE)  
BIOCOMPOSITES

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## ABSTRACT

Pure cellulose was dissolved in 7 wt.% sodium hydroxide (NaOH)/12 wt.% urea at the temperature of -12 °C to prepare the regenerated cellulose (RC). Structural, morphological and thermal properties of cellulose and RC were compared using field emission scanning electron microscopy (FESEM), Fourier transform infrared spectroscopy (FTIR), x-ray diffraction (XRD), thermogravimetric analysis (TGA) and differential scanning calorimetry (DSC). The results revealed decreased in crystallinity, reduced of FTIR peak intensity and decreased the thermal stability of RC sample. FESEM results showed that the morphological of RC became more spherical. RC/poly(3-hydroxybutyrate) (PHB) biocomposites were prepared by melt mixing in internal mixer with different RC content (1, 3, 5 and 7 wt.%). The structural, morphological, thermal and mechanical properties of the RC/PHB were characterized. From the XRD, the crystallinity was decreased with the increasing of RC fiber loading. The melting temperature of biocomposites was found to increase upon the increasing of RC fiber. TGA results indicated that the char residue and onset temperature of biocomposites were decreased with the increasing of RC loading. Increased loading of RC decreased the tensile strength and elongation at break, while the Young's modulus showed the optimum value at 5 wt.%. Based on flexural strength and flexural modulus, 3 wt.% RC was the optimum amount for PHB composite. As for comparison of PHB composite with 3 wt.% cellulose and 3 wt.% RC, DSC result showed the melting point of 3 wt.% RC/PHB composite was higher than 3 wt.% cellulose/PHB composite. The tensile properties and impact strength were higher for cellulose/PHB compared to RC/PHB composites, while flexural properties of RC/PHB were higher than cellulose/PHB composites.

## ABSTRAK

Selulosa tulen dilarutkan di dalam larutan akueus 7 wt.% natrium hidroksida (NaOH) / 12 wt.% urea pada suhu -12 °C untuk menyediakan selulosa terjanasemula (RC). Struktur, morfologi dan sifat haba selulosa dan RC dibandingkan menggunakan mikroskop elektron imbasan pancaran medan (FESEM), spektroskopi inframerah transformasi Fourier (FTIR), pembelauan sinar-x (XRD), analisis termogravimetri (TGA) dan kalorimetri imbasan pembezaan (DSC). Keputusan sample RC menunjukkan penurunan kehabluran, penurunan intensiti puncak FTIR dan penurunan kestabilan haba. Keputusan FESEM menunjukkan bahawa morfologi RC menjadi lebih berbentuk sfera. RC/poli(3-hidroksibutirat) (PHB) biokomposit telah disediakan dengan pencampuran leburan dengan kandungan RC yang berbeza (1, 3, 5 dan 7 wt.%) dalam pengadun. Struktur, morfologi, sifat haba, sifat mekanik RC/PHB telah dicirikan. Daripada keputusan XRD, kehabluran telah menurun dengan peningkatan gentian RC. Suhu lebur biokomposit didapati meningkat dengan penambahan gentian RC. Keputusan TGA menunjukkan sisa hangus dan suhu permulaan RC/PHB telah menurun dengan peningkatan kandungan RC. Peningkatan RC telah menurunkan sifat kekuatan tegangan dan pemanjangan pada takat putus, manakala modulus Young menunjukkan nilai optimum pada 5 wt.% gentian RC. Berdasarkan kekuatan lenturan dan modulus lenturan, 3 wt.% RC adalah nilai optimum bagi komposit PHB. Dalam perbandingan antara 3 wt.% selulosa dan 3 wt.% RC, kajian DSC menunjukkan takat lebur komposit 3 wt.% RC/PHB adalah lebih tinggi, berbanding komposit 3 wt.% selulosa/PHB. Sifat tegangan dan kekuatan impak komposit selulosa/PHB adalah lebih tinggi daripada komposit RC/PHB, manakala sifat lenturan komposit RC/PHB adalah lebih tinggi daripada komposit selulosa/PHB.

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**LIST OF ABBREVIATIONS**

AR	-	Analytical Reagents
ASTM	-	American Society for Testing and Materials
BCNW	-	Bacterial Cellulose Nanowhiskers
CSF	-	Cellulose Standard Fiber
CNF	-	Cellulose Nano Fiber
NaOH	-	Sodium Hydroxide
CS <sub>2</sub>	-	Carbon Disulfide
DTG	-	Derivative Thermogravimetry
DSC	-	Differential Scanning Calorimetry
FTIR	-	Fourier Transform Infrared Spectroscopy
H <sub>2</sub> SO <sub>4</sub>	-	Sulfuric Acid
H <sub>2</sub> O	-	Water
IC	-	Inclusion Complex
LCCE	-	Long Chain Cellulose Ester
MCCF	-	Microcrystallite Cellulose Fiber
Na <sub>2</sub> SO <sub>4</sub>	-	Sodium Sulfate
OH group	-	Hydroxyl Group
phr	-	Part per hundred resin
PHA	-	Polyhydroxyalkanoates

PHB	-	Poly(3-hydroxybutyrate)
PHBV	-	Poly(3-hydroxybutyrate-co-3-hydroxyvalerate)
PP	-	Polypropylene
rpm	-	Rotation per minute
PCL	-	Poly( $\epsilon$ -caprolactone)
PLA	-	Poly(lactic Acid)
SEM	-	Scanning Electron Microscope
TGA	-	Thermogravimetric Analyse
wt. %	-	Weight percentage
XRD	-	X-ray Diffractometer

**LIST OF SYMBOLS**

$F$	-	Force
$g$	-	Gravity = 9.81 m/s
$p$	-	Pressure
$\rho$	-	Density
$T$	-	Temperature

## CHAPTER 1

### INTRODUCTION

#### 1.1 Background of Research

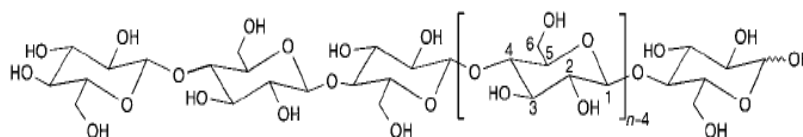
Polymer composites are mixtures of polymer matrix with additives, which will form two or more phases and components ((Xanthos *et al.*, 2010). There are two types of additives, which are organic and inorganic, where involving much geometry such as spheres, fibers, particulates and flakes.

The development of commercial petroleum based synthetic polymer beneficial for decades. However, it causes severe problems like environmental pollution and over exploitation of fossil resources. The most possible way to develop raw materials that environmentally compatible is by preparing the biodegradable biopolymers (Hatakeyana *et al.*, 2002). Nowadays, there are many research using renewable resource natural fiber in the polymer composites as a reinforcements. These bring benefits to the environment with respect to the biodegradable and utilization of natural materials (Rout *et al.*, 2001).

Poly(3-hydroxybutyrate) (PHB) is one of the biopolymers, which produced by saturated aliphatic polyester. It can be synthesized by variety type of bacteria as a metabolism of stored energy molecule (Mokhtari-Hosseini *et al.*, 2009). PHB is a semicrystalline polymers that biodegradable and has biocompatibility. PHB is mainly used in biomedical field and helping to relieve environmental pollution due to the disposal of non biodegradable petroleum based polymer (Savenkova *et al.*, 2000).



Cellulose is a semicrystalline polysaccharide which has large amount of hydroxyl groups that responsible for the hydrophilic properties of the natural fibers (Schuster *et al.*, 2004). It is the basic building block in most of the plants (Krassig *et al.*, 1993). Cellulose is a carbohydrate polymer generated from the repeating  $\beta$ -D-glucopyranose molecular units that are covalently linked between the equatorial OH group of C1 carbon atom and C4 carbon atom. Cellulose is an extensive and linear-chain polymer, consists a large number of hydroxy functional groups. Chain length of cellulose determined by the degree of polymerization, depend on the treatment and origin of the raw material. Structure of cellulose chain consists a D-glucose unit (C4-OH) at one end, while the other end is C1-OH group (Klemm *et al.*, 2005a).



**Figure 1.1:** Molecular structure of cellulose (n= degree of polymerisation)(Klemm *et al.*, 2005a)

Due to renewable and environmentally friendly biomaterial, plant cellulose contributes about  $2 \times 10^{12}$  tons of the annually biomass production, which are photosynthesized by the fixation of carbon dioxide in sea and land (Klemm *et al.*, 2005a). The advantages of the used of cellulose applications are the hydrophilicity, biocompatibility and the production of cellulose composite with either biopolymer or synthetic polymers (Kim *et al.*, 2006). It also exhibited wide applications. Cellulose is biodegradability, abundance, stiffness and high mechanical strength. Besides this, cellulose also exhibit low weight and price, and the characteristics such as a very large surface-to-volume ratio and outstanding electrical, mechanical and thermal properties thus became the most abundant renewable biopolymer material (Gabr *et al.*, 2014).

Cellulose powder used as raw material in different type of applications including packaging, drugs, paints and toothpaste. In its original form, cellulose is

classified as the cellulose I, which also called native cellulose. After undergo mechanical or chemical treatment, the cellulose is classified as cellulose II, which also called as regenerated cellulose (RC) (Kompella and Lambros, 2002). Crystalline structure of native cellulose exist as cellulose I, formed by two polymorphs: triclinic (Ia) and monoclinic (Ib) which appear alongside each other. Varies origin of cellulose have different Ia/Ib.

Cellulose application and development are limited due to its difficulty to dissolve in most of the common solvents due to its strong inter-molecular force, van der Waals and intra-molecular force of hydrogen bonds (Heinze and Koschella, 2005). Other than that, the cellulose also does not melt, making it difficult to be processed using common processing method. Moreover, traditional technologies that produce RC materials are not environmental friendly causing serious environmental problem and pollution. Therefore, processing approaches that prevent toxic byproducts and complicated processing routes is important (Guansen *et al.*, 2012).

Sodium hydroxide (NaOH)/urea aqueous solvent system have been reported as a good alternatif and green solvent of cellulose. NaOH and urea are inexpensive and nontoxic chemical agents. NaOH treatment or dissolution of the cellulose followed by precipitation can be used to form cellulose II. Cellulose II have an antiparallel orientation, in contrast to native cellulose I which have parallel chains. This conversion cannot be reversed, indicating that cellulose II is stable (Osullivan *et al.*, 1997). There is no evaporation of any toxic byproducts at the time of cellulose dissolution at low temperature. During the commercialize and large scale production, this aqueous system can maintain “green” atmosphere air in the surrounding environment. The byproducts also can be easily recycled or separated. (Qi *et al.*, 2009).

## 1.2 Problem Statement

Recently, there is a great interest in the study of PHB biopolymers, driven by its biodegradability however it has limited range of applications due to its high production cost and low mechanical properties. Due to high crystallinity, PHB is brittle and stiff thus reduced the mechanical properties of PHB with a low extension at break, which limit the use of PHB (Seggiani *et al.*, 2015).

Cellulose is abundant and relatively cheap resources. Cellulose fibers have the advantages of renewable, nontoxic, biodegradable and easily obtained from natural raw materials. Their commercial usage has been limited because they have lower thermal, mechanical and gas barrier properties if compared to synthetic polymers. However, cellulose can be incorporated into polymer matrix to enhance thermal and mechanical properties. Cheng *et al.* (2007) studied the lyocell regenerated cellulose fiber and microfibrillated cellulose to reinforce polyvinyl alcohol. The tensile modulus found to improve as increasing amount of lyocell and microfibrillated cellulose fiber, especially at filler content of in between 2% to 6%.

In the research of Viviana *et al.* (2009), the double layer of PHB composite with acetylated cellulose cardboard found to have a better interfacial adhesion compared to the PHB/pure cellulose cardboard composites. This is due to diminish of hydrophilic properties of cellulose, and increased in interfacial adhesion with PHB. Mechanical properties of polymer composites can be affected depending on the cellulose treatment. In the research of Dormann *et al.* (2016) on all-cellulose composite laminates, with short dissolution time in the aqueous of 7 wt.% NaOH/12 wt.% urea of 5 min led to improvement in tensile strength of 114 MPa and a Young's modulus of 7.8 GPa. Thus, NaOH/urea treatment has potential to develop an environmental friendly fiber biocomposite. Through the regeneration process, the morphology of RC fiber would change thus improving mechanical and thermal properties due to higher interfacial adhesion of RC fiber to PHB matrix as compared to cellulose fiber.

This study focused on preparation of RC and the effects of RC loading on PHB properties. RC was prepared using NaOH/urea aqueous solution due to simple preparation, economical, environmental friendly and fast dissolubility. Effect of the RC fiber loading ratio in PHB matrix was investigated, in order to find the optimum loading ratio of RC fibers on thermal, mechanical, structural and morphology properties of composites.

### **1.3 Objective**

The overall objective is to develop a biocomposites based on PHB/RC.

The specific objectives are:

1. To prepare the RC via NaOH/urea aqueous and characterize the morphological and thermal properties.
2. To investigate the effects of RC content on the morphology, mechanical and thermal properties of PHB/RC biocomposites.
3. To compare the properties of PHB composites of 3 wt% of cellulose and RC.

### **1.4 Scopes of the Study**

The scope of study includes the following criteria:

- (i) From the purchased cellulose powder, the RC fiber were prepared using urea/ NaOH regeneration process.

- (ii.) The RC fiber were mixed with PHB in internal mixer. Then, the compound were hotpress in compression machine into suitable thickness for mechanical testing.
- (iii.) The sample were cut into suitable dimension, then were characterized using:
- a. Thermogravimetric Analyser (TGA)  
TGA was used to measure thermal stability of the material. The tests were performed non-isothermally at constant heating rate to obtain the onset of thermal degradation and char residue.
  - b. Differential Scanning Calorimetry (DSC)  
DSC testing was used to measure the melting temperature, glass transition temperature, crystallization temperature and crystallinity.
  - c. Tensile testing  
Tensile testing was one of the mechanical testing. The samples were subjected to a tensile force in constant force. Values of tensile strength, tensile modulus and elongation at break were obtained.
  - d. Flexural testing.  
Flexural testing method was used to measure behavior of materials when subjected to three points loading. Flexural strength, and flexural modulus were provided.
  - e. Izod Impact Tester.  
The Izod impact tester was used to determine the impact resistance and amount of energy that absorbed by material in order to fracture. Notch was cut across the testing sample. Pendulum hammer was release and hit the sample to break it. Impact energy was recorded.

f. Field Emission Scanning Electron Microscopy (FESEM)

FESEM gives the high magnification and resolution images by focus the electron beam across material surface. It is used to examine the interface, size and surface of the materials.

g. X-ray Diffractometer (XRD)

The percentage crystallinity in the material was identified.

h. Fourier Transform Infrared Spectroscopy (FTIR)

FTIR was used to identify the chemical functional group in material.

## REFERENCE

- Abdelwahab, M. A., Flynn, A., Chiou, B., Imam, S., Orts, W. and Chiellini, E. (2012). Thermal, Mechanical and Morphological Characterization of Plasticized PLA and PHB Blends. *Polymer Degradation And Stability*. 97(9), 1822-1828.
- Adusumali, R. B., Reifferscheid, M., Weber, H., Roeder, T., Sixta, H. and Gindl, W. (2006). Mechanical Properties of Regenerated Cellulose Fibres for Composites. *Macromolecule Symposium*, 244(1),119–125.
- Ahmed Mohamed El-Hadi (2013). Influence of Microcrystalline Cellulose Fiber (MCCF) on the Morphology of Poly(3-Hydroxybutyrate) (PHB). *Colloid Polymer Science*. 291(3), 743–756.
- Alata, H., Hexig, B. and Inoue, Y. (2006). Effect of Poly(vinyl alcohol) Fine Particles as a Novel Biodegradable Nucleating Agent on the Crystallization of Poly(3-hydroxybutyrate). *Journal Polymer Science Part B Polymer Physic*. 44(13),1813-1820.
- Anderson, A. J. and Dawes, E. A. (1990). Occurrence, Metabolism, Metabolic Role, and Industrial Uses of Bacterial Polyhydroxyalkanoates. *Microbiological Reviews*. 54(4), 450-472.
- Arrakhiz, F.Z., El Achaby, M., Malha, M., Bensalah, M.O., Fassi-Fehri, O., Bouhfid, R., Benmoussa, K. and A. Qaiss (2013). Mechanical and thermal properties of natural fibers reinforced polymer composites: Doum/low density polyethylene. *Materials & Design*. 43, 200-205.
- Avella, M., Martuscelli, E., Pascucci, P., Raimo, M., Focher, B. and Marzetti, A. (1993). A New Class of Biodegradable Materials: Poly-3-hydroxybutyrate/Steam Exploded Straw Fiber Composites: Thermal and Impact Behaviour. *Journal of Applied Polymer Science*. 49(12), 2091-2103.
- Averous, L. and Boquillon, N. (2004). Biocomposites based on Plasticized Starch: Thermal and Mechanical Behaviours. *Carbohydrate Polymer*. 56: 111–122. Biopolymer And Sustainable Raw Material. *Angewandte Chemie. International Edition*. 44(22), 3358–3393.

- Benmoussa, K. and A. Qaiss (2013). Mechanical and thermal properties of natural fibers reinforced polymer composites: Doum/low density polyethylene. *Materials & Design*. 43, 200-205.
- Boissou, F., Muhlbauer, A., Vigier, K. D. O., Leclercq, L., Kunz, W., Marinkovic, S., Estrine, B., Rataj, V. N. and Jerome, F. (2014). Transition of cellulose crystalline structure in biodegradable mixtures of renewably-sourced levulinate alkyl ammonium ionic liquids,  $\gamma$ -valerolactone and water. *Green Chemistry*. 16(5), 2463–2471.
- Bouafif, H., Koubaa, A., Perre, P. and Cloutier, A. (2009). Effects Of Fiber Characteristics On The Physical And Mechanical Properties Of Wood Plastic Composites. *Composites: Part A*. 40(12), 1975–1981.
- Brandl, H., Gross, R. A., Lenz, R. W. and Fuller, R. C. (1990). Plastics from Bacteria and for Bacteria: Poly(Beta-Hydroxyalkanoates) as Natural, Biocompatible, and Biodegradable Polyesters. *Advances in Biochemical Engineering/Biotechnology*. 41, 77–93.
- Burgaz, E., Yazici, M., Kapusuz, M., Alisir, S. H. and Ozcan, H. (2014). Prediction of thermal stability, crystallinity and thermomechanical properties of poly(ethylene oxide)/clay nanocomposites with artificial neural networks. *Thermochimica Acta*. 575, 159–166.
- Cai, J. and Zhang, L. (2005). Rapid Dissolution of Cellulose In LiOH/ Urea and NaOH/Urea Aqueous Solutions. *Macromolecule Bioscience*. 5(6), 539–548.
- Cai, J. and Zhang, L. (2006). Unique Gelation Behavior of Cellulose in NaOH/Urea Aqueous Solution. *Biomacromolecules*. 7(1), 183.
- Cai, J., Wang, L. and Zhang, L. (2007a). Influence of Coagulation Temperature on Pore Size and Properties of Cellulose Membranes Prepared From NaOH–Urea Aqueous Solution. *Cellulose*. 14(3), 205–215.
- Cai, J., Zhang, L., Zhou, J., Qi, H., Chen, H., Kondo, T., Chen, X. and Chu, B. (2007b). Multifilament Fibers Based on Dissolution of Cellulose in NaOH/Urea Aqueous Solution: Structure And Properties. *Advance Material*. 19(6), 821.
- Cao, Y., Li, H., Zhang, Y., Zhang, J. and He, J.(2010). Structure and Properties of Novel Regenerated Cellulose Films Prepared from Cornhusk Cellulose in Room Temperature Ionic Liquids. *Journal of Applied Polymer Science*. 116(1), 547–554.



- Capadona, J. R., Shanmuganathan, K., Tyler, D. J., Rowan, S. J., and Weder, C. (2008). Stimuli-Responsive Polymer Nanocomposites Inspired by the Sea Cucumber Dermis. *Science*. 319(5868), 1370–1374.
- Carrilo, F., Colom, X., Sunol, J. J. and Saurina, J. (2004). Structural FTIR analysis and thermal characterization of lyocell and viscose-type fibres. *Europe Polymer Journal*. 40(9): 2229-2234.
- Chan, R. T. H., Garvey, C. J., Marcal, H., Russell, R. A., Holden, P. J. H. and Foster, L. J. R. (2011). Manipulation of Polyhydroxybutyrate Properties through Blending with Ethyl-Cellulose for a Composite Biomaterial. *International Journal of Polymer Science*. 2011(651549), 1-8.
- Chang, C., Chen, S. and Zhang, L. J. (2011). Novel Hydrogels Prepared via Direct Dissolution of Chitin at Low Temperature: Structure and Biocompatibility. *Material Chemistry*. 21(11), 3865-3871.
- Chen, H. Z., Wang, N. and Liu, L. Y. (2012). Regenerated cellulose membrane prepared with ionic liquid 1-butyl-3-methylimidazolium chloride as solvent using wheat straw. *Journal Chemical Technology Biotechnology*. 87(2), 1634–1640.
- Cheng, Q., Wang, S., Rials, T. G. and Lee, S. (2007). Physical and Mechanical Properties of Polyvinyl Alcohol and Polypropylene Composite Materials Reinforced with fibril Aggregates Isolated From Regenerated Cellulose fibers. *Cellulose*. 14(6), 593–602.
- Christian, S. J. and Billington, S. L. (2011). Mechanical Response of PHB and Cellulose Acetate Natural Fiber-reinforced Composites for Construction Applications. *Composites: Part B*. 42(7), 1924.
- Ciolacu, D., Ciolacu, F. and Popa, V. I. (2011). Amorphous Cellulose – Structure And Characterization. *Cellulose Chemistry And Technology*. 45 (1-2), 13-21.
- Dimonie1, M. and Rapa, M. (2010). Biodegradable Blends based on PHB and Wood Fiber. *U.P.B. Science Bull, Series B*. 72(3), 6-7.
- Dormanns, J. W., Schuermann, J., Mussig, J., Duchemin, B. J. C. and Staiger, M. P. (2016). Solvent Infusion Processing Of All-Cellulose Composite Laminates Using And Aqueous NaOH/urea Solvent System. *Composites: Part A*. 82, 130–140.

- Egal, M., Budtova, T. and Navard, P. (2008). The Dissolution of Microcrystalline Cellulose in Sodium Hydroxide-Urea Aqueous Solutions. *Cellulose*. 15(3), 361–370.
- El-Hadi, A., Schnabel, R., Straube, E., Muller, G. and Henning, S. (2002). Correlation between Degree of Crystallinity, Morphology, Glass Temperature, Mechanical Properties and Biodegradation of poly (3-hydroxyalkanoate) PHAs and their Blends. *Polymer Test*. 21(6), 665-674.
- Elanthikkal, S., Gopalakrishnapanicker, U., Varghese, S., James, T. G. and Francis, T. (2013). Effect of cellulose whisker content on the properties of poly(ethylene-co-vinyl acetate)/cellulose composites. *Carbohydrate Polymers*. 95, 773–779.
- Elashmawi, I. S. and Hakeem, N. H. (2014). Production and modification of PEO/PVDF Membranes Loaded with Silver Nitrate. *Research Journal of Pharmaceutical, Biological and Chemical Sciences*. 5(4), 1356-1367.
- Furukawa, T., Sato, H., Murukami, R., Zhang, J., Duan, Y., Noda, I., Ochiai, S. and Ozak, Y. (2005). Structure, Dispersibility, and Crystallinity of Poly(hydroxybutyrate)/Poly(l-lactic acid) Blends Studied by FT-IR Microspectroscopy and Differential Scanning Calorimetry. *Macromolecules*. 38(15), 6445-6454.
- Gabr, M. H., Okubo, K. and Fujii, T. (2014). Mechanical and Morphology Properties of Cellulose Nanocomposites. Pandey, J.K., Takagi, H., Nakagaito, A.N., Kim, H.-J. (Eds.) Volume C: Polymer Nanocomposites of Cellulose Nanoparticles Handbook of Polymer Nanocomposites. Processing, Performance and Application. (pp. 249-263). London: Springer-Verlag Berlin Heidelberg.
- Graupner, N., Holger, F., Gerhard, Z. and Jorg, M. (2014). Improvement and Analysis of Fibre/Matrix Adhesion of Regenerated Cellulose Fibre Reinforced PP-, MAPP- and PLA-Composites by the Use of Eukalyptus Globulus Lignin. *Composites: Part B*. 66, 117-125.
- Gunning, M. A., Geever, L. M., Killion, J.A., Lyons, J. G. and Higginbotham, C.L. (2013). Mechanical and Biodegradation Performance of Short Natural Fibre Polyhydroxybutyrate Composites. *Journal Polymer Testing*. 32(8), 1603–1611.

- Han, J., Zhou, C., D. French, A., Han, G. and Wu, Q. (2013). Characterization of cellulose II nanoparticles regenerated from 1-butyl-3-methylimidazolium chloride. *Carbohydrate Polymers*. 94 (2013), 773– 781.
- Hatakeyama, H. (2002). Thermal Analysis of Environmentally Compatible Polymer Containing Plant Components in the Main Chain. *Polymer Advance Technology*. 70(3): 151–155.
- Heinze, T. and Koschella, A. (2005). Solvents Applied in the Field of Cellulose Chemistry: A Mini Review. *Polimeros*. 15(2), 84–90.
- Higgins, H.G., Goldsmith, V. and Mckenzie, A. W.(1958). The Reactivity Of Cellulose Iii, Acid Hydrolysis Of Eucalypt A-Cellulose In The Intermediate Molecular Weight Range. *Journal of Polymer Science*. 32, 57-74.
- Hocking, P. J., Marchessault, R. H. and Griffin, G. J. L. (1995). Chemistry and Technology of Biodegradable Polymers. *Journal of Chemical Education*. 72 (3), A73.
- Hon, D. N. S. (1996). Functional Polymers: a New Dimensional Creativity in Lignocellulosic Chemistry. Hon, D. N. S. (Ed.) In *Chemical Modification of Lignocellulosic Materials*(pp. 1-10). New York: Marcel Dekker Incorporation.
- Ho, M., Wang, H., Lee, J., Ho, C., Lau, K., Leng, J. and Hui, D. (2011). Critical Factors On Manufacturing Processes Of Natural Fibre Composites. *Composites: Part B*. 43(8), 3549–3562.
- Hu, F., Lin, N., Peter, R. and Huang, J. (2015). Reinforcement and nucleation of acetylated cellulose nanocrystals in foamed polyester composites. *Carbohydrate Polymers*. 129, 208–215.
- Huan, S., Bai, L., Cheng, W. and Han, G. (2016). Manufacture of electrospun all-aqueous poly(vinyl alcohol)/cellulose nanocrystal composite nanofibrous mats with enhanced properties through controlling fibers arrangement and microstructure. *Polymer*. 92,25-35.
- Iglesias, M. E. P., Urzua, L. S., Flores, M. L. O. F., Velazquez, D. L. and Morales, L. J. M. (2014). Extraction and NMR Determination of PHB from *Azospirillum brasilense* Sp7. *Journal of Chemical, Biological and Physical Sciences*. 4 (5), 26-32.
- Janoobi, M., Harun, J., Mathew, A. P., Oksman, K. (2010). Mechanical Properties of Cellulose Nanofiber (CNF) Reinforced Polylactic Acid (PLA) Prepared by

- Twin Screw Extrusion. *Composites Science And Technology*. 70(12), 1742–1747.
- Josepha, P.V., Joseph, K., Thomas, S., Pillai, C. K. S., Prasad, V.S., Groeninckx, G. and Sarksova, M. (2003). The Thermal And Crystallisation Studies Of Short Sisal Fibre Reinforced Polypropylene Composites. *Composites: Part A*. 34(3), 253–266.
- Khalil, H. P. S. A., Bhat, A. H. and Yusra, A. F. I. (2012). Green Composites from Sustainable Cellulose Nanofibrils: A Review. *Carbohydrate Polymers*. 87(2), 963-979.
- Kargarzadeh, H., Sheltami, R. M., Ahmad, I., Abdullah, I. and Dufresne, A. (2015). Cellulose Nanocrystal: A Promising Toughening Agent For Unsaturated Polyester Nanocomposite. *Polymer*. 56, 346-357.
- Kim, J., Song, C., and Yun, S. (2006). Cellulose based Electro-active Paper: Performance and Environmental Effects. *Smart Materials and Structure*. 15(3), 719–725.
- Kim, G. M., Lach, R., Michler, G. H., Potschke, P. and Albrecht, K. (2006). Relationships Between Phase Morphology And Deformation Mechanisms In Polymer Nanocomposite Nanofibres Prepared By An Electrospinning Process. *Nanotechnology*. 17(4), 963-972.
- Klemm, D., Brigitte, H., Hans-Peter, F. and Andreas, B. (2005a). Cellulose: Fascinating Biopolymer and Sustainable Raw Material. *Angewandte Chemie International Edition. Polymer science*. 44(22), 3358 – 3393.
- Klemm, D., Heublein, B., Fink, H. P. and Bohn, A. (2005b). Cellulose: Fascinating Renewable Resources: Opportunities and Challenges in the Green Materials World. *Journal of Polymers and the Environment*. 10(1/2), 19–26.
- Kompella, M. K. and Lambros, J. (2002). Micromechanical Characterization of Cellulose Fibers. *Polymer Testing*. 21(5), 523–530.
- Kowalczyk, M., Piorkowska, E., Kulpinski, P. and Pracella, M. (2011). Mechanical and Thermal Properties of PLA Composites With Cellulose Nanofibers and Standard Size Fibers. *Composites: Part A*. 42(10), 1509–1514.
- Krassig, H. A. (1994). Cellulose: Structure, Accessibility and Reactivity. *Journal of Polymer Science Part A: Polymer Chemistry*. 32(12), 2401.
- Krishnaprasad, R., Veena, N. R., Hanna, J. M., Rathish, R., Mikael, S. and Kuruvilla, J. (2009). Mechanical and Thermal Properties of Bamboo Microfibril

- Reinforced Polyhydroxybutyrate Biocomposites. *Journal Polymer Environment*. 17(2), 109–114.
- Lee, S. Y. and Choi, J. (2001). Production of Microbial Polyester by Fermentation of Recombinant Microorganisms. *Advances in Biochemical Engineering/Biotechnology*. 71, 183–207.
- Lenz, R. W. and Marchessault, R. H. (2005). Bacterial Polyesters: Biosynthesis, Biodegradable Plastics and Biotechnology. *Biomacromolecule*. 6(1), 1-8.
- Lin, N., Chen, Y., Hu, F. and Huang, J. (2015). Mechanical Reinforcement Of Cellulose Nanocrystals On Biodegradable Microcellular Foams With Melt-Compounding Process. *Cellulose*. 22, 2629–2639.
- Liu, Z., Wang, H., Li, Z., Lu, X., Zhang, X., Zhang, S., Zhou, K. (2011). Characterization Of The Regenerated Cellulose Films In Ionic Liquids And Rheological Properties Of The Solutions. *Materials Chemistry and Physics*. 128(1-2), 220–227.
- Luengo, J. M., Garcia, B., Sandoval, A., Naharo, G. and Olivera, E. R. (2003). Bioplastics from microorganisms. *Current Opinion in Microbiology*. 6(3), 251–260.
- Macedo, J. d. S., Costa, M. F., Tavares, M. I. B. and Thire, R. M. S. M. (2010). Preparation and Characterization of Composites Based on Polyhydroxybutyrate and Waste Powder from Coconut Fibers Processing. *Polymer Engineering And Science*. 50 (7) , 1466–1475.
- Mariotti, N., Wang, X., Rodrigue, D. and Stevanovic, T. (2014). Combination of Esterified Kraft Lignin and MAPE as Coupling Agent for Bark/HDPE Composites. *Journal of Materials Science Research*. 3(2), 8-22.
- Martinez, S. M., Villano, M., Oliveira, C., Albuquerque, M. G., Majone, M., Reis, M., Lopez, R. A. and Lagaron, J. M. (2014). Characterization of Polyhydroxyalkanoates Synthesized from Microbial Mixed Cultures and of their Nanobiocomposites with Bacterial Cellulose Nanowhiskers. *New Biotechnology* . 31(4), 364-376.
- Mary, S. K., Sasidharan Pillai, P. K., Amma, D. B., Pothan, L. A. and Thomas, S. (2013). Aging and Biodegradation of Biocomposites. Thomas, S., Durand, D., Chassenieux, C. and Jyotishkumar, P.(Ed.). *Handbook of Biopolymer-Based Materials: From Blends and Composites to Gels and Complex Networks* (pp.

- DOI: 10.1002/9783527652457.ch26). Weinheim, Germany : Wiley-VCH Verlag GmbH & Co. KGaA.
- Melo, J. D. D., Carvalho, L. F. M., Medeiros, A. M., Souto, C.R.O. and Paskocimas, C.A. (2012). A Biodegradable Composite Material Based on Polyhydroxybutyrate (PHB) and Carnauba Fibers. *Composites: Part B*. 43(7), 2827–2835.
- Mirjalili, F., Chuah, L. and Salahi, E. (2014). Mechanical and Morphological Properties of Polypropylene/Nano  $\alpha$ -Al<sub>2</sub>O<sub>3</sub> Composite. *Scientific World Journal*. Doi: 10.1155/2014/718765.
- Miranda, M.I.G., Bica, C.I.D., Nachtigall, S.M.B., Rehman, N. and Rosa, S.M.L. (2013). Kinetical thermal degradation study of maize straw and soybean hull celluloses by simultaneous DSC–TGA and MDSC techniques. *Thermochimica Acta*. 565, 65–71.
- Mokhtari-Hosseini, Z. B., Vasheghani-Farahani, E., Heidarzadeh-Vazifekhoran, A., Shojaosadati, S. A., Karimzadeh, R., Khosrav, D. K. (2009). Statistical Media Optimization for Growth and PHB Production from Methanol by a Methylophilic Bacterium. *Bioresource Technology*. 100(8), 2436–2443.
- Mousarioum, P., Halley, P. J. and Doherty, W. O. S. (2013). Thermophysical Properties and Rheology Of PHB/Lignin Blends. *Industrial Crops and Products*. 50, 270–275.
- Nabi, S. D. and Jog, J. P. (1999). Natural Fiber Polymer Composites: A Review. *Advances in Polymer Technology*. 18, 351–363.
- Nadhan, A. V. and Rajulu, A. V. (2012). Properties of Regenerated Cellulose Short Fibers/Cellulose Green Composite Films. *Journal of Polymer Environment*. 20(2), 454–458.
- Nassima, E. M., Karima, A., Abdellatif, B., Mohamed, Z., Aziz, F., Abderrahim, S., and Mounir, E. Achaby., (2015). Bio-nanocomposite films reinforced with cellulose nanocrystals: Rheology of film-forming solutions, transparency, water vapor barrier and tensile properties of films. *Carbohydrate Polymers*. 129,156–167.
- Nawrath, R. and Poirier, N. (2008). Pathways for the Synthesis of Polyesters in Plants: Cutin, Suberin, and Polyhydroxyalkanoates. *Advances In Plant Biochemistry And Molecular Biology*. 1, 201-239.

- Nawrath, C., Poirier, Y. and Somerville, C. (1994). Targeting of the Polyhydroxybutyrate Biosynthetic Pathway to the Plastids of Arabidopsis Thaliana Results in High Levels of Polymer Accumulation. *Proceedings Of The National Academy Of Sciences*. 20 December. USA, 91: 12760–12764.
- Nejad, M. H. (2011). Improving the Mechanical Properties of Polysaccharide Derivatives through Melt Compounding with Nano-Clays. Doctor Philosophy. Technische University Berlin, German.
- Osullivan, A. C. (1997). Cellulose: The Structure Slowly Unravels. *Cellulose*. 4(3), 173-207.
- Peng, Y., Gardner, D. J. and Yousoo, H. (2015). Characterization of mechanical and morphological properties of cellulose reinforced polyamide 6 composites. *Cellulose*. 22(31), 99-321.
- Pereira, P. H. F., Voorwld, H. J. C., Cioffi, M. O. H. and Silva, M. L. C. P. D. (2011). Novel Cellulose/NbOPO<sub>4</sub>.nH<sub>2</sub>O Hybrid Material From Sugarcane Bagasse. *BioResources*. 6(1), 867-878.
- Qi, H., Cai, J. and Zhang, L. (2009). Properties Of Films Composed of Cellulose Nanowhiskers and a Cellulose Matrix Regenerated from Alkali/Urea Solution. *Biomacromolecules*. 10(6), 1597–1602.
- Qi, H., Chang, C. and Zhang, L. (2009). Properties and Applications of Biodegradable Transparent and Photoluminescent Cellulose Films Prepared via a Green Process. *Green Chemistry*. 11(2), 177–184.
- Qin, X., Lu, A. and Zhang, L. (2012). Effect of Stirring Conditions on Cellulose Dissolution in NaOH/Urea Aqueous Solution at Low Temperature. *Journal Of Applied Polymer Science*. 126 (S1), E470–E477.
- Ramamoorthy, S. K., Kundu, C. K., Adekunle, K., Bashir, T. and Skrifvars. M. (2013). Properties of Green Composites with Regenerated Cellulose Fiber and Soybean-Based Thermoset for Technical Applications. *Journal of Reinforced Plastics and Composites*. 0(00), 1–9.
- Rapa, M., Popa, M. E., Cornea, P. C., Popa, V.L., Grosu, E., Geicu-Cristea, M., and Petruta Stoica, P. (2014). Degradation study by trichoderma spp. of poly (3-hydroxybutyrate) and wood fibers composites. *Romanian Biotechnological Letters*. 19 (3), 9390-9399.
- Rathanak, P., Idayu, M. I. and Razak, R. A. (2005). Mechanical Properties And Biodegradability Study Of Coconut Shell Powder (CSP) Filled Low Density

- Polyethylene (LDPE) Film. *Simposium Polimer Kebangsaan Ke-V*. 23-24 Ogos 2005. Hotel Residence: *Simposium Polimer Kebangsaan*. 2005. 257-267.
- Ray, D., Sarkar, B. K., Basak, R. K. B. and Rana, A. K. (2004). Thermal behavior of vinyl ester resin matrix composites reinforced with alkali-treated jute fibers. *Journal of Applied Polymer Science*. 94(1), 123-129.
- Repaske, R. and Repaske, A. C. (1976). Quantitative Requirements for Exponential Growth of *Alcaligenes Eutrophus*. *Applied Environmental Microbiology*. 32(4), 585-591.
- Reza, A., Azman, H., Haafiz, M. K. M. and Zainoha, Z. (2015). Effect of Microcrystalline Cellulose on Biodegradability, Tensile and Morphological Properties of Montmorillonite Reinforced Polylactic Acid Nanocomposites. *Fibers and Polymers*, 16(10), 2284-2293.
- Robles, E., Urruzola, I., Labidi, J. and Serrano, L. (2015). Surface-modified nanocellulose as reinforcement in poly(lactic acid) to conform new composites. *Industrial Crops and Products*. 71, 44-53.
- Rojas, J. (2013). Effect of Polymorphism on the Particle and Compaction Properties of Microcrystalline Cellulose. Ven, T. V. D. and Godbout, L. (Eds.). *Cellulose - Medical, Pharmaceutical and Electronic Applications* (pp. 27-46). Croatia: InTech.
- Rojo, E., Alonso, V., Oliet, M., Saz-Orozco, B. D. and Rodriguez, F. (2015). Effect of fiber loading on the properties of treated cellulose fiber-reinforced phenolic composites. *Composites: Part B* (68), 185-192.
- Rosa, D. I. M., Santulli, C. and Sarasini, F. (2010). Mechanical and Thermal Characterization of Epoxy Composites Reinforced with Random and Quasi-Unidirectional Untreated Phormium Tenax Leaf Fibers. *Materials And Design*. 31(5), 2397-405.
- Rosa, S. M. L., Santos, E. F., Ferreiral, C. A. and Nachtigall, S. M. B. (2009). Studies on the properties of rice-husk-filled-PP composites - effect of maleated PP. *Material Research*. 12(3), 333-338.
- Rout, J., Misra, M., Tripathy, S., Nayak, S. K. and Mohanty, A. K. (2001). The Influence of Fibre Treatment on the Performance of Coir-Polyester Composites. *Composite Science Technology*. 61(9), 1303-1310.
- Rowell, R. M. (1995). A New Generation of Composite Materials from Agro-Based Fiber. Fai, T. J., Mark., James., Prasad, E. and Paras, N. (Eds.). *Polymers and*



- other Advanced Materials: Emerging Technologies and Business Opportunities. (pp. 659-665). New York: Springer US.
- Rusli, R., Shanmuganathan, K., Rowan, S. J., Weder, C. and Eichhorn, S. J. (2010). Stress-Transfer in Anisotropic and Environmentally Adaptive Cellulose Whisker. *Biomacromolecules*. 11(3), 762-768.
- Sanadi, A. R., Caulfield, D. F., Jacobson, R. E. and Rowell, R. M.(1995). Renewable Agricultural Fibers as Reinforcing Fillers in Plastics: Mechanical Properties of Kenaf Fiber-Polypropylene Composites. *Industrial & Engineering Chemistry Research*. 34(5), 1889-1896.
- Savenkova, L., Gercberga, Z., Nikolaeva, V., Dzene, A., Bibers, I., and Kalnin, M. (2000). Mechanical Properties and Biodegradation Characteristics of PHB-based Films. *Process Biochemistry*. 35(6), 573–579.
- Scandola, M., Ceccorulli, G., & Pizzoli, M. (1992). Miscibility of bacterial poly(3hydroxybutyrate) with cellulose esters. *Macromolecules*. 25, 6441–6446.
- Schuster, K. C., Rohrer, C., Eichinger, D., Schmidtbauer, J., Aldred, P. and Firgo, H. (2004). Environmentally Friendly Lyocell Fibers. Wallenberger, Frederick, T. and Weston, N. (Ed.) Natural Fibers, Plastics and Composites. (pp. 123-146). New York: Kluwer Academic Publishers.
- Segal, L., Creely, J. J., Martin, A. E., and Conrad, C.M. (1959). An empirical method for estimating the degree of crystallinity of native cellulose using the X-Ray diffractometer. *Textile Research Journal*. 29, 786–794.
- Seggiana, M., Cinelli, P., Verstichelc, S., Puccinia, M., Vitoloa, S., Anguillesia, I. and Lazzeri, A. (2015). Development of Fibres-Reinforced Biodegradable Composites. *Chemical Engineering Transactions*. 43, 1813-1818.
- Shibata, S., Cao, Y. and Fukumot, I. (2005). Effect Of Bagasse Fiber On The Flexural Properties Of Biodegradable Composites. *Journal of polymer composite*. 26(5), 689-694.
- Shi, Q. F., Mou, H. Y., Gao, L., Yang, J., and Guo, W. H. (2010). Double-Melting Behavior of Bamboo Fiber/Talc/Poly (Lactic Acid) Composites. *Journal of Polymers and The Environment*. 18(4),567–575.
- Srithep, Y., Ellingham, T., Peng, J., Sabo, R., Clemons, C., Turng, L. and Pilla, S. (2013). Melt Compounding of Poly (3-Hydroxybutyrate-Co-3-

- Hydroxyvalerate)/Nanofibrillated Cellulose Nanocomposites. *Polymer Degradation and Stability*. 98(8), 1439-1449.
- Steinbuechel, A. and Valentin, H. E. (1995). Diversity of Bacterial Polyhydroxyalkanoic Acids. *FEMS Microbiology Letters*, 128 (3), 219–228.
- Suttiwijitpukdee, N., Sato, H., Zhang, J., Hashimoto, T. and Ozaki, Y. (2011). Intermolecular interactions and crystallization behaviors of biodegradable polymer blends between poly (3-hydroxybutyrate) and cellulose acetate butyrate studied by DSC, FT-IR, and WAXD. *Polymer*. 52, 461-471.
- Tsekos, I. (1999). The Sites of Cellulose Synthesis in Algae: Diversity and Evolution of Cellulose Synthesizing Enzyme Complexes. *Journal of Phycoogy*, 35, 635–655.
- Verhoogt, H., Ramsay, B. A. and Favis, B. D. (1994). Polymer Blends Containing Poly(3-hydroxyalkanoate)s. *Polymer*. 35(24), 5155-5169.
- Verhoogt, H., Ramsay, B. A., Favis, B. D. and Ramsay, J. A. (1996). The Influence of Thermal History on the Properties of Poly(3-Hydroxybutyrate-Co-12%-3-Hydroxyvalerate). *Journal. Applied Polymer Science*. 61(1), 87-96.
- Viviana, P. C., Commisso, M. S. and Vazquez, A. (2009). Biocomposites Based on Renewable Resource: Acetylated and Non Acetylated Cellulose Cardboard Coated with Polyhydroxybutyrate. *Polymer*. 50(26), 6274–6280.
- Ward, A. C., Rowley, B. I. and Dawes, E. A. (1977). Effect of Oxygen and Nitrogen Limitation on Poly-β-Hydroxybutyrate Biosynthesis in Ammonium-Grown *Azotobacter Beijerinckii*. *Journal of General Microbiology*. 102, 61-68.
- Xanthos, M. (2010). Polymers and polymer composites. Xanthos, M. (Ed.) *Functional Fillers for Plastics*. ( pp. 1-18 ) Weinheim: Verlag GmbH & Co. KGaA.
- Xu, Y., Wang, C., Stark, N. M., Cai, Z. and Chu, F. (2012). Miscibility and Thermal Behavior of Poly (β-Caprolactone)/Long-Chain Ester of Cellulose Blends. *Carbohydrate Polymers*. 88, 422– 427.
- Yamane, C., Saito, M., Okajima, K. (1996). Specification of Alkali Soluble Pulp Suitable for New Cellulosic Filament Production. *Sen-I Gakkaishi*. 52(6),318–324
- Yan, L., Chen, J. and Bangal, P. R. (2007). Dissolving Cellulose in a NaOH/Thiourea Aqueous Solution: A Topochemical Investigation. *Macromolecule Bioscience*. 7(9-10), 1139–1148.

- Yan, L., Tao, H., Bangal, P. R. C. (2009). Synthesis and Flocculation Behavior of Cationic Cellulose Prepared in a NaOH/Urea Aqueous Solution. *Clean–Soil, Air, Water*. 37 (1), 39-44.
- Yang, Z., Xu, S., Ma, X. and Wang, S. (2008). Characterization and Acetylation Behavior of Bamboo Pulp. *Wood Science Technology*. 42(8), 621–632.
- Yu, H., Liu, R. G., Shen, D. W., Jiang, Y. & Huang, Y. (2005). Study on Morphology and Orientation of Cellulose in the Vascular Bundle of Wheat Straw. *Polymer*. 46, 5689–5694.
- Yu, H. Y., Qin, Z. Y., Liu, N. Y., Chen, L., Liu, N. and Zhou, Z. (2012). Simultaneous Improvement of Mechanical Properties and Thermal Stability of Bacterial Polyester by Cellulose Nanocrystals. *Carbohydrate Polymers*. 89(3), 973-974.
- Zeronian, S. H. and Ryu, H. S. (1987). Properties of cotton fibers containing the cellulose IV crystal structure. *Journal Applied Polymer Science*. 33(7), 2587–2604.
- Zhang, H., Wu, J., Zhang, J. and He, J. (2005). 1-allyl-3-methylimidazolium chloride room temperature ionic liquid: a new and powerful nonderivatizing solvent for cellulose. *Macromolecules*. 38 (20), 8272–8277.
- Zhang, Y. H. and Lynd, L. R. (2004). Toward an aggregated understanding of enzymatic hydrolysis of cellulose: noncomplexed cellulase systems. *Biotechnology Bioengineering*. 88(7):797-824.
- Zhao, H., Kwak, J., Wang, Y., Franz, J. A. , White, J. M. and Holladay, J. E. (2007). Interactions Between Cellulose and N-Methylmorpholine-N-Oxide. *Carbohydrate*. 67(1), 97–103.