UNIVERSITI PUTRA MALAYSIA

DEVELOPMENT OF POLY(LACTIC ACID)/KENAF-DERIVED CELLULOSE WITH THERMALLY GRAFTED AMINOSILANE AND EPOXIDIZED PLANT OILS FOR POTENTIAL FOOD PACKAGING APPLICATIONS

TEE YEE BOND

FK 2015 1

DEVELOPMENT OF POLY(LACTIC ACID)/KENAF-DERIVED CELLULOSE WITH THERMALLY GRAFTED AMINOSILANE AND EPOXIDIZED PLANT OILS FOR POTENTIAL FOOD PACKAGING APPLICATIONS

By

TEE YEE BOND

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

June 2015

Abstract of thesis presented to the Senate of Universiti Putra Malaysia in fulfilment of the requirement for the degree of Doctor of Philosophy

DEVELOPMENT OF POLY(LACTIC ACID)/KENAF-DERIVED CELLULOSE WITH THERMALLY GRAFTED AMINOSILANE AND EPOXIDIZED PLANT OILS FOR POTENTIAL FOOD PACKAGING APPLICATIONS

By

TEE YEE BOND

June 2015

Chair: Rosnita A. Talib, PhD

Faculty: Engineering

Poly(lactic acid) (PLA) biocomposites aimed for food packaging applications were developed from the present research. Seventeen PLA blends were studied and one optimum ternary biocomposite was finalized to conclude the work. Kenaf-derived cellulose (C), 3-aminopropyltriethoxysilane (APS), epoxidized soybean oil (ESO), and epoxidized palm oil (EPO) were the core materials tested for reinforcement.

It is typical that natural fibers-filled plastic composites to report poor interfacial adhesion due to their distinct difference in polarity. To improve the interfacial adhesion, kenaf-derived cellulose was treated with silane coupling agent, APS prior to filling into PLA. Moreover, they were subjected to thermal treatment for permanent cross-linking upon acknowledging that desorption of chemicals is highly undesirable in food-related packaging. Also distinct to typical practices, C was thermally grafted with various silane concentrations (5, 8, and 11 wt%) and these silane-grafted cellulose (SGC) were termed as SGC5, SGC8, and SGC11. From investigation via Fourier transform infrared spectroscopy (FTIR), thermogravimetric analysis (TGA), morphology analysis, size distribution, and density measurement, it was conclusive that (i) cellulose was derived from kenaf, and (ii) silane was effectively grafted onto the cellulose.

30 wt% of C, SGC5, SGC8, and SGC11 were respectively melt-compounded into PLA and hot-pressed into 0.3-mm sheets with the aim to develope biocomposite with optimum properties. With highest reinforcement in tensile strength, water resistance, dimensional stability, crystallinity, and oxygen barrier, PLA/SGC8 was concluded as the biocomposite for development towards packaging applications. Nonetheless, all biocomposites did not improve PLA's brittle nature. Concurrently, they reduced the toughness. With sustainability in mind, EPO and commercialized ESO were plasticizers considered to reinforce these properties. Out of the analyses subjected, it is noteworthy that EPO can transition PLA to become ductile and tougher at lower concentration than ESO which denoted its competency as alternative bioplasticizer.

Comprehending EPO's potential as reactive plasticizer, the research continued with EPO addition at various loadings (3, 5, and 10 wt%) into PLA/SGC8 composite. Peak shifts were observed in the FTIR spectra of PLA/SGC8/EPO blends which was absent in PLA/C/EPO. It was proposed that EPO could reactively interact with PLA and SGC which therefore functioned as a compatible layer between the interfaces. The proposed mechanism was validated from morphology analysis plus improvement in water resistance, mechanical, and thermal properties of PLA/SGC8/EPO blends. Of all ternary blends, PLA/SGC8/EPO5 reported optimum reinforcement in EAB and toughness. The oxygen and water vapour barrier of this optimized blend suggested potential as packaging material for salad mixes.

Conceding that the developed biocomposite can be impractical if it cannot be fabricated into functional product, PLA/SGC8/EPO5 together with PLA/EPO10, PLA/C/EPO5, and PLA (as reference) sheets were selectively subjected to thermoformability and overall migration test. While PLA/C/EPO5 failed both tests, PLA/SGC8/EPO5 was thermoformable and constituent migration was within the stipulated overall migration limit. These practical tests reasserted the competency of the presently developed biocomposite to be fabricated towards food packaging applications.

Abstrak tesis yang dikemukakan kepada Senat Universiti Putra Malaysia
sebagai memenuhi keperluan untuk ijazah Doktor Falsafah

PEMBANGUNAN POLI(ASID LAKTIK)/SELULOSA TERBITAN KENAF YANG DIGRAFKAN SECARA TERMA DENGAN SILANA-AMINO DAN MINYAK TUMBUHAN YANG DIEPOKSIKAN UNTUK POTENSI APLIKASI PEMBUNGKUSAN MAKANAN

Oleh

TEE YEE BOND

Ogos 2015

Pengerusi: Rosnita A. Talib, PhD

Fakulti: Kejuruteraan

Biokomposit poli(asid laktik) (PLA) untuk aplikasi pembungkusan makanan telah dibangunkan dalam penyelidikan ini. Daripada tujuh belas campuran PLA yang dikaji, satu biokomposit dengan sifat-sifat optimum telah dimuktamadkan. Selulosa terbitan kenaf (C), '3-aminopropyltriethoxysilane' (APS), minyak kacang soya yang diepoksikan (ESO), dan minyak kelapa sawit yang diepoksikan (EPO) adalah bahan utama yang dipilih untuk pengukuhan biokomposit.

Sering kali, komposit plastik yang diisi dengan fiber semulajadi mempunyai lekatan antara muka yang lemah atas perbezaan ionik yang ketara. Untuk mengatasi masalah tersebut, selulosa terbitan kenaf dirawat dengan silana APS sebelum diisi ke dalam PLA. Selain itu, mereka turut diberi rawatan terma supaya silana-amino digrafkan secara kekal ke atas C. Hal demikian kerana lesapan kimia adalah amat tidak diingini dalam aplikasi pembungkusan makanan. Juga berbeza dengan amalan tipikal, C digrafkan secara terma dengan silana-amino pada pelbagai kepekatan (5, 8, dan 11%) dan mereka dinamakan sebagai SGC5, SGC8, dan SGC11. Daripada kajian Spektroskopi Inframerah Fourier (FTIR), analisis Termogravimetri (TGA), analisis morfologi, taburan saiz, dan pengukuran ketumpatan, adalah disahkan bahawa (i) selulosa berjaya diterbitkan daripada kenaf, dan (ii) silana digrafkan ke atas selulosa dengan berkesan.

30% C, SGC5, SGC8, dan SGC11 dicampur ke dalam PLA dan ditekan secara terma untuk penghasilan kepingan komposit dengan ketebalan 0.3-mm. Ia bertujuan untuk membangunkan biokomposit dengan ciri-ciri optimum. Dengan peningkatan optimum dalam kekuatan tegangan, rintangan air, kestabilan dimensi, penghabluran, dan rintangan oksigen, PLA/SGC8 disimpulkan sebagai biokomposit untuk pembangunan ke arah aplikasi pembungkusan. Walau bagaimanapun, semua biokomposit tidak meningkatkan keboleh muluran PLA. Malah, kekuatan impaknya merosot. Disebabkan kemapanan sumbernya, EPO dan ESO yang telah dikomersialkan dipertimbangkan sebagai pemplastik *(plasticizer)* untuk mengukuhkan sifat-sifat tersebut. Daripada analisis-analisis yang dijalankan, EPO boleh meningkatkan kemuluran dan kekuatan impak PLA pada kepekatan yang lebih rendah daripada ESO. Hal demikian mengesahkan kecekapan EPO sebagai bio-pemplastik alternatif.

Setelah memahami potensi EPO sebagai pemplastik reaktif, penyelidikan diteruskan dengan penambahan EPO pada pelbagai kepekatan (3, 5, dan 10%) ke dalam komposit PLA/SGC8. Terdapat perubahan puncak dalam spektrum FTIR campuran PLA/SGC8/EPO yang tidak wujud dalam spektrum FTIR PLA/C/EPO. Justeru itu, adalah dicadangkan bahawa EPO boleh berinteraksi secara reaktif dengan PLA dan SGC. EPO mungkin boleh berfungsi sebagai lapisan keserasian antara PLA dan SGC. Mekanisme interaksi yang dicadangkan boleh disahkan daripada analisis morfologi serta peningkatan ketara pada rintangan air, mekanikal, dan sifat-sifat terma dalam campuran PLA /SGC8/EPO. Daripada sampel-sampel komposit yang dikaji, PLA/SGC8/EPO5 melaporkan peningkatan optimum kemuluran dan kekuatan impak. Daripada penyelidikan rintangan oksigen dan wap air, adalah dicadangkan bahawa biokomposit tersebut berpotensi untuk digunakan dalam pembungkusan campuran salad.

Atas pemahaman bahawa biokomposit yang dibangunkan boleh menjadi tidak praktikal jika ia tidak boleh dibentuk ke dalam produk berfungsi, PLA/SGC8/EPO5 bersama-sama dengan PLA, PLA/EPO10, dan PLA/C/EPO5 dipilih untuk ujian pembentukan plastik secara terma dan penghijrahan keseluruhan bahan. PLA/C/EPO5 gagal dalam ujian-ujian tersebut. Sementara itu, PLA/SGC8/EPO5 boleh dibentuk secara terma dengan baik dan penghijrahan bahan daripadanya berada dalam had penghijrahan yang dibenarkan. Ujian-ujian praktikal tersebut memberi sokongan kukuh ke atas kesesuaian biokomposit yang kini dibangunkan untuk difabrikasi ke arah aplikasi pembungkusan makanan.

ACKNOWLEDGEMENTS

I would like to express my sincerest gratitude to my supervisor, Dr. Rosnita A. Talib who has unconditionally supported and guided me throughout to ensure the well going of this project. Her encouragements, constructive criticisms, and of course the time spent in reviewing my work is very much appreciated. I would also like to thank my co-supervisors, namely Assoc. Prof. Ir. Dr. Chin Nyuk Ling, Dr. Khairul Faezah Md. Yunos, and Assoc. Prof. Khalina Abdan for the advices and constructive alternatives given in upholding the quality of the present work.

I am thankful to the staffs for their assistance, service, and experience shared during my research work at the laboratories and workplaces. They were from Department of Process and Food Engineering (UPM), Department of Chemical and Environmental Engineering (UPM), Laboratory of Biocomposite Technology and Laboratory of Biopolymer and Derivatives at Institute of Tropical Forestry and Forest Products (INTROP, UPM), Halal Products Research Institute (UPM), Radiation Processing Technology Division of Malaysian Nuclear Agency (MNA), Packaging Research Centre Sdn Bhd, and Mecomb Malaysia Sdn Bhd. I also acknowledge Miss Patpen Penjumras, Miss Intan Syafinaz, and the rest of the researchers whom I have made friends for the time, encouragement, and knowledge shared.

Last but not least, I owe a big thanks to my family especially my parents for their loving support, patience, and faith in me in pursuing this postgraduate study. For that, I dedicate this dissertation to my beloved parents, Tee Chong Seng and Yeow Meng Choo.

I certify that a Thesis Examination Committee has met on 30th June 2015 to conduct the final examination of Tee Yee Bond on his thesis entitled "Development of Poly(Lactic Acid)/Kenaf-Derived Cellulose with Thermally Grafted Aminosilane and Epoxidized Plant Oils for Potential Food Packaging Applications" 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 Doctor of Philosophy.

Members of the Thesis Examination Committee were as follows:

Mohd Nordin bin Ibrahim, PhD
Associate Professor
Faculty of Engineering
Universiti Putra Malaysia
(Chairman)

Russly bin Abdul Rahman, PhD
Professor
Faculty of Engineering
Universiti Putra Malaysia
(Internal Examiner)

Yus Aniza binti Yusof, PhD
Associate Professor
Faculty of Engineering
Universiti Putra Malaysia
(Internal Examiner)

Rafael Auras, PhD
Associate Professor
School of Packaging
Michigan State University
United States
(External Examiner)

ZULKARNAIN ZAINAL, PhD
Professor and Deputy Dean
School of Graduate Studies
Universiti Putra Malaysia

Date: 12 August 2015

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

Rosnita A. Talib, PhD
Senior Lecturer
Faculty of Engineering
Universiti Putra Malaysia
(Chairman)

Chin Nyuk Ling, PhD
Associate Professor, Ir.
Faculty of Engineering
Universiti Putra Malaysia
(Member)

Khairul Faezah Md. Yunos, PhD
Senior Lecturer
Faculty of Engineering
Universiti Putra Malaysia
(Member)

Khalina Abdan, PhD
Associate Professor
Faculty of Engineering
Universiti Putra Malaysia
(Member)

BUJANG KIM HUAT, 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.: ____

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: ______________________

Signature: ______________________
Name of Member of Supervisory Committee: ______________________

Signature: ______________________
Name of Member of Supervisory Committee: ______________________

Signature: ______________________
Name of Member of Supervisory Committee: ______________________

TABLE OF CONTENTS

LIST OF TABLES

LIST OF FIGURES

LIST OF ABBREVIATIONS

AGM	Acetyl glycerol monolaurate
ANOVA	Analysis of Variance
AOTD	Advance Oleochemical Technology Division
APS	3-aminopropyl triethoxysilane
ASTM	American Society of Testing and Materials
ATBC	Acetyl(tributyl citrate)
BBP	Butylbenzylphthalate
C	Cellulose
CAS	Chemical Abstract Service
CES	Cyanoethyltrimethoxy silane
COPP	γ-(2,3-cyclopropoxy)-propyltrimethoxy Silane
DBP	Di-n-butylphthalate
DBS	Dibutyl sebacate
DEHP	Di-(2-ethylhexyl) phthalate
DOA	Bis(2-ethylhexyl) adipate
DOP	Dioctyl phthalate
DSC	Differential scanning calorimetry
DTG	Differential Thermogravimetry
ECER	East Corridor Economic Region
EHO	Epoxidized hemp oil
ELO	Epoxidized linseed oil

EPO	Epoxidized palm oil
ESO	Epoxidized soybean oil
FCM	Food contact material
FCS	Food contact substance
FTIR	Fourier transform infrared
GPa	Giga Pascal
GPS	3-glycidoxypropyltrimethoxysilane
GRAS	Generally recognised as safe
GTA	Glyceryl triacetate
HAP	Hydroxyapatite
HDI	Hexamethylenediisocyanate
HIPS	High-impact polystyrene
HPMC	Hydroxypropyl methylcellulose
ICKAF	International Conference on Kenaf and Allied Fibers
INTROP	Institute of Tropical Forestry and Forest Product
ISO	International Organization for Standardization
KBF	Kenaf bast fiber
LDPE	Low density polyethylene
MA	Maleic anyhidride
MAP	Modified atmosphere packaging
MCC	Microcrystalline cellulose
MFC	Microfibrillated cellulose

MGST	Maleic anhydride-grafted starch
MMT	Sodium montmorillonite
MPa	Mega Pascal
MPOB	Malaysian Palm Oil Board
MPS	3-methacryloxypropyltrimethoxysilane
NaOH	Sodium hydroxide
OLA	Oligomeric lactic acid
OML	Overall migration limit
OPEFB	Oil palm empty fruit bunch
OTR	Oxygen transmission rate
PBOH	Poly(1,3-butanediol)
PBS	Poly(butylenes succinate)
PCL	Polycaprolactone
PE	Polyethylene
PEG	Poly(ethylene glycol)
PET	Poly(ethylene terephthalate)
PF	Phenolformaldehyde
PHA	Polyhydroxyalkanoates
PHB	Poly(hydroxylbutyrate)
PHBV	Poly(hydroxybutyrate-valerate)
PLA	Poly(lactic acid)
PP	Polypropylene
PPG	Poly(propylene glycol)
PS	Polystyrene

PVC	Polyvinyl carbonate
RSF	Rice straw fiber
SGC	Silane-grafted cellulose
SML	Specific migration limit
TBAC	Tributyl O-acetylcitrate
TEC	Triethyl citrate
T_g	Glass transition temperature
TGA	Thermogravimetric Analysis
TOR	Threshold of Regulation
USFDA	United States Food and Drug Administration
v%	Volume fraction
VPSEM	Vapour Pressure Scanning Electron Microscopy
VTS	Vinyltrimethoxysilane
wt%	Weight Fraction
WVTR	Water vapour transmission rate

CHAPTER 1

INTRODUCTION

1.1 Statement and Significance of the Problem

There is always a vast interest in developing new bio-based products. The cumulative concern from the depleting petroleum, the increasing social and environmental concern, and the stringent government regulations especially in Asian and European countries have made the need for “greener” products more vital than ever. The packaging industry is among the most obvious target for these efforts as the usage of packaging materials has shown continuous increase over time (Johansson *et al*., 2012).

In terms of plastic production, the technology development of petroleum-based plastics are more long-standing as compared to the latterly developing bioplastics such as polyhydroxylalkanoates (PHAs) and poly(lactic acid) (PLA). Mechanical and physical performance limitations as well as high costs from low-volume production became barriers for the widespread acceptance of these bioplastics (Mohanty *et al.,* 2000). To empower these biomaterials as the wave of the future, the incorporation of inexpensive natural fibers can be done in which they do not only decrease in production cost, but they may offer additional sustainability advantages.

Acknowledging that PLA made the utmost impact in the packaging industry and as much as 60 % of packaging was used in the food and beverage sector, it is aimed to develop a PLA biocomposite suitable for food packaging applications (Johansson *et al*., 2012). When food related packaging is in concern, it is crucial to incorporate only substances that are approved for food contact and personal care. For safety measures, in addition to cross-checking with the international standards for approved chemical substances, it is decided to emphasize on the usage of bio-sourced materials in the present composite development. Cellulose was derived from local crop kenaf and was added into PLA as fillers. However, one major drawback of using natural fibers as composite fillers is their marked hydrophilicity nature and highly polar character. This limits their compatibility with the polymeric matrices which are mostly hydrophobic and non-polar (Xie *et al*., 2010). Among other chemical treatments, many researchers treated the natural fibers with silane coupling agent to improve the filler-matrix interfacial adhesion. Similar treatment was also applied in the present work. However, foreseeing the possible reverse hydrolysis of the chemical compound and leaching from the cellulose fillers which are highly undesirable in food packaging, it was brought a step further with thermal grafting to permanently cross-link the silanol and siloxane polymer networks onto the cell walls. The effectiveness of the thermal treatment and its influence on the fillers' and composites' characteristics were discussed.

Another concern with PLA is the brittle nature which limits its' processability and the range of product development. Addition of silanized-cellulose could have increased the tensile strengths and moduli of the PLA; however it does not contribute in toughening it. Researchers have used plasticizers such as triacetin and poly(ethylene gycol) (PEG) onto PLA/natural fibers composites (Ibrahim *et al*., 2010; Masirek *et al*., 2007). However, these manmade plasticizers were unable to provide PLA/natural fibers composites an effective brittle-toughness transition. Recently, several authors have tested plant oils as plasticizers for PLA/starch composites and reported possible reaction between the modified starch and the plant oils and toughen the composites (Xiong *et al.,* 2014; 2013c; 2013b; 2013a). Acknowledging that this could be a new path for fabricating an entirely bio-sourced composite, the present composites were plasticized with a novel local product, which is the epoxidized palm oil. Considering the compatibility and the ability of epoxidized palm oil as a reactive plasticizer for the PLA/silanized-cellulose composites, enhancement in the overall properties of the biocomposites is anticipated.

Generally with the right technique and combinations, PLA/natural fiber composites with good improvements can be achieved. However, there must be a critical cost-efficiency consideration. It is acknowledged that natural fiber composites will succeed more in the markets when they cost less than conventional composites for similar resolution (Thamae and Baillie, 2007). For the current research, with extraction of cellulose method adapted from standards similar to extracting cellulose from wood for pulp and paper production, using a comparatively small concentration of silane coupling agent to treat the fillers, and the use of sustainable and safe plant oil as reactive plasticizers, it is envisage that the novel biocomposite engineered can be economically viable.

1.2 Research Aim and Objectives

The aim of this research is to modify cellulose that was derived from kenaf bast fibers and use epoxidized palm oil as a novel plasticizer for poly(lactic acid) (PLA)/kenaf-derived cellulose biocomposite reinforcement to widen its window of application specifically in packaging food products. To achieve the aim, several objectives were set as listed below:

1 To improve the compatibility between the kenaf-derived cellulose and PLA via thermally grafting coupling agent 3-aminopropyltriethoxy silane (APS) onto cellulose.

2 To evaluate the plasticizing effect of epoxidized soybean oil (ESO) and epoxidized palm oil (EPO) on PLA matrix.

3 To determine the reinforcing mechanism(s) involved with various loadings of silane used to treat kenaf-derived cellulose and various loadings of EPO added in PLA/kenaf-derived cellulose composites.

4 To evaluate EPO as a novel reactive plasticizer in the PLA/kenaf-derived cellulose composite system.

5 To characterize the fillers and composites in terms of chemical, mechanical, physical, thermal, barrier, and morphological properties.

1.3 Significance of the Research

1 A novel research on the interaction between plant-derived cellulose that was silane-grafted thermally and bio-sourced plastic, PLA.

2 Testing the potential of Malaysia's bio-sourced product, EPO as an alternative plasticizer.

3 A novel study on the interaction mechanism involved for EPO to reinforce the properties of PLA/kenaf-derived cellulose composites.

4 Development of a novel biocomposite suitable for food packaging applications.

1.4 Thesis Outline

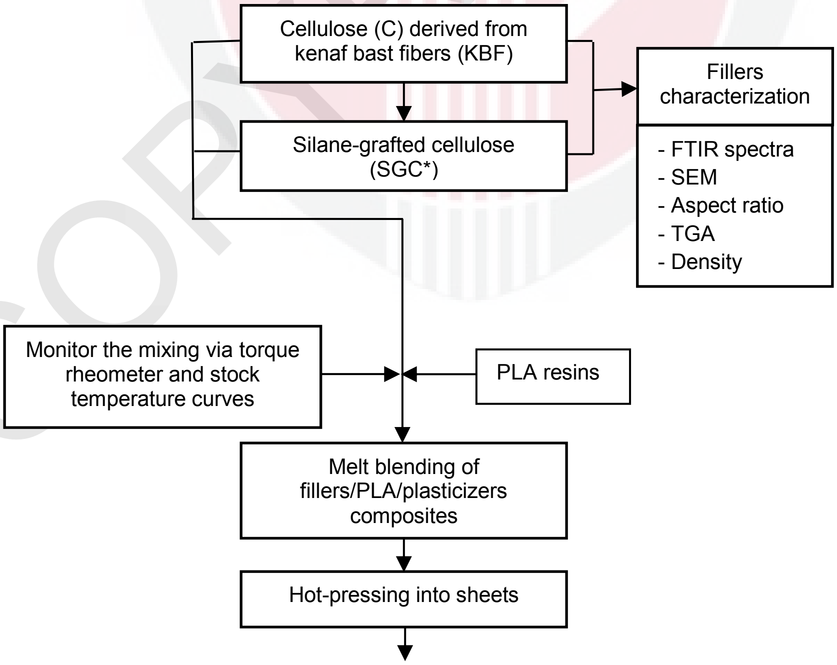
The thesis consists of 6 chapters. Chapter 1 served as an introduction to the research which includes the statement and significance of problem, aim and objectives, as well as the significance of the research.

Chapter 2 presented the literature review on PLA polymer, natural fibers and their modifications, plasticizers, plastic packaging, and composites development in various areas. This chapter also reviewed previous findings on plastic-related composites in terms of chemical, mechanical, physical, thermal, barrier, and morphological properties.

Chapters 3 to 5 are the working chapters where the methodologies, results, and discussions were conveyed accordingly to the objectives set in Setion 1.2. The objectives in Chapter 3 were (i) to improve compatibility between kenaf-derived cellulose and PLA via thermal grafting aminosilane onto the cellulose prior to filling; followed by (ii) the determination of the reinforcing mechanism involved via various loadings of silane used for treatment. Analyses on properties of kenaf bast fibers (KBF), kenaf-derived cellulose (C), and silane-grafted cellulose (SGC) at various silane wt% were done. This was followed with the studies of binary composites, namely PLA/C and all PLA/SGC blends. To conclude, a PLA/SGC formulation with optimum characteristics was chosen for further modification as continued in Chapter 4.

Chapter 4 initiated with the evaluation of epoxidized palm oil (EPO) as alternative bio-plasticizer via the comparison with PLA plasticized with commercialised epoxidized soybean oil (ESO). This was followed by the addition of EPO at various loadings into PLA/SGC composites to evaluate the reinforcing mechanisms among the constituents. To conclude, a PLA/SGC/EPO formulation with optimum characteristics was determined.

As the final working chapter, Chapter 5 presented important practical evaluation on the optimized PLA/SGC/EPO blends along with some of the prepared samples in terms of their thermoformability and overall migration from the blend. Their applicability and suitability for food packaging were discussed. Finally, Chapter 6 presented the overall conclusions of the research and recommendations for future research. The process flow chart for the research methodology is as shown in Figure 1.1.



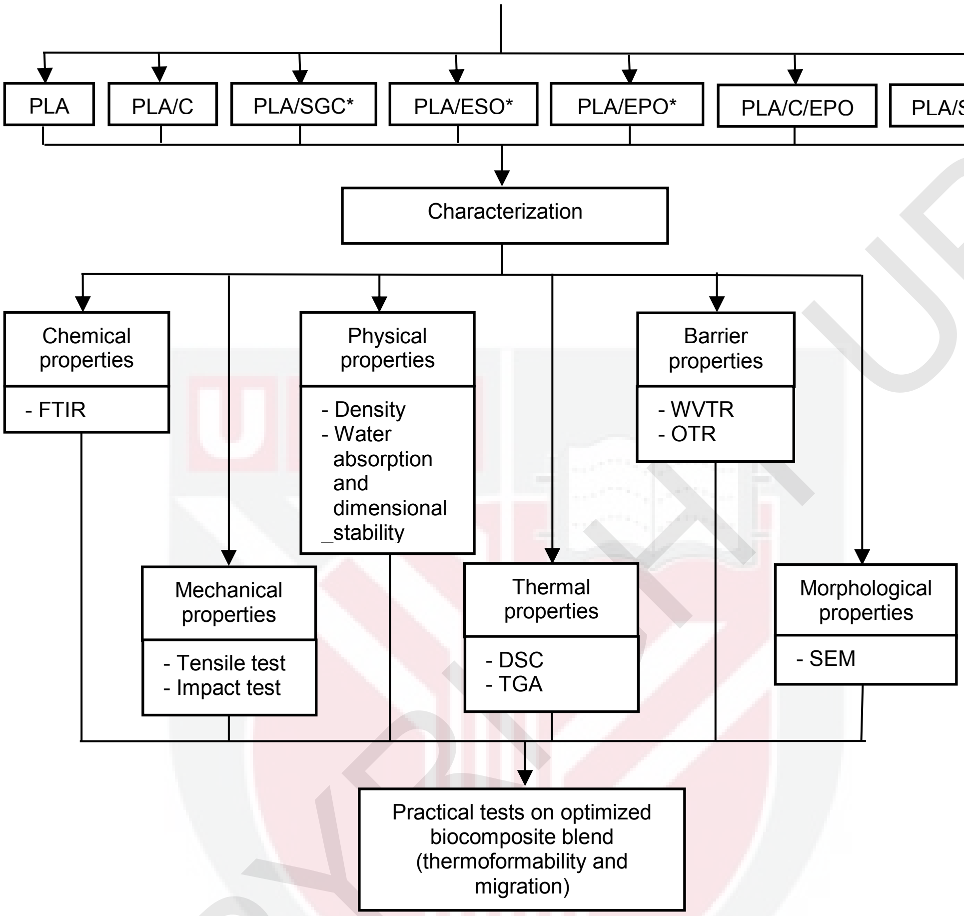


Figure 1.1: Process flow chart *(*more than one formulation)*.

REFERENCES

21 C. F. R. 170.39. *Food and Drugs (Threshold of regulation for substances used in food-contact articles)*. Code of Federal Regulations. U.S. Food and Drug Administration. Revised as of April 1, 2014. Title 21, Volume 3.

21 C. F. R. 184. *Food and Drugs (Direct food substances affirmed as generally recognized as safe)*. Code of Federal Regulations. U.S. Food and Drug Administration. Revised as of April 1, 2014. Title 21, Volume 3, Part 184.

21 C. F. R. 186. *Food and Drugs (Indirect food substances affirmed as generally recognized as safe)*. Code of Federal Regulations. U.S. Food and Drug Administration. Revised as of April 1, 2014. Title 21, Volume 3.

21 C. F. R. 175.300. *Food and Drugs (Resinous and polymeric coatings)*. Code of Federal Regulations. U.S. Food and Drug Administration. Revised as of April 1, 2014. Title 21, Volume 3.

21 C. F. R. 178.3297. *Food and Drugs (Colorants for polymers).* Code of Federal Regulations. U.S. Food and Drug Administration. Revised as of April 1, 2014. Title 21, Volume 3.

Abbott, S. 2010. Chemical compatibility of poly(lactic acid): A practical framework using Hansen Solubility Parameters. In *Poly(lactic acid): Synthesis, Structures, Properties, Processing, and Applications (2nd ed.)*, eds. R. Auras, L. T. Lim, S. E. M. Selke, and H. Tsuji, pp. 83-95. Hoboken, New Jersey, US: John Wiley & Sons, Inc..

Abdan, K., Harun, J., and Aw Yeong, C. H. 2008. Properties of kenaf (*Hibiscus Cannabinus L.*) bast fibre reinforced unsaturated polyester composite. *In Research in Natural fibre reinforced polymer composites*, ed. S. Salit, pp. 233-246. Malaysia: UPM Publisher.

Abdellatief, A. and Welt, B. A. 2012. Comparison of new dynamic accumulation method for measuring oxygen transmission rate of packaging against the steady-state method described by ASTM D3985. *Packaging Technology and Science* 26(5): 281-288.

Abdelmouleh, M., Boufi, S., Belgacem, M. N., and Dufresne, A. 2007. Short natural-fibre reinforced polyethylene and natural rubber composites: Effect of silane coupling agents and fibres loading. *Composites Science and Technology* 67(7): 1627-1639.

Abdelmouleh, M., Boufi, S., Belgacem, M. N., Duarte, A. P., Salah, A. B., and Gandini, A. 2004. Modification of cellulosic fibres with functionalised silanes: development of surface properties. *International Journal of Adhesion and Adhesives* 24(1): 43-54.

Abdelmouleh, M., Boufi, S., ben Salah, A., Belgacem, M. N., and Gandini, A. 2002. Interaction of silane coupling agents with cellulose. *Langmuir* 18(8): 3203-3208.

Abdullah, N. M., and Ahmad, I. 2012. Effect of chemical treatment on mechanical and water-sorption properties coconut fiber-unsaturated polyester from recycled PET. *ISRN Materials Science* 2012: 1-8.

Adhvaryu, A., and Erhan, S. Z. 2002. Epoxidized soybean oil as a potential source of high-temperature lubricants. *Industrial Crops and Products* 15(3): 247-254.

Ahmad, S. H., Rasid, R., Bonnia, N. N., Zainol, I., Mamun, A. A., Bledzki, A. K., and Beg, M. D. H. 2011. Polyester-kenaf composites: Effects of alkali fiber treatment and toughening of matrix using liquid natural fiber. *Journal of Composite Materials* 45(2): 203-217.

Aji, I. S., Sapuan, S. M., Zainudin, E. S., and Abdan, K. 2009. Kenaf fibres as reinforcement for polymeric composites: A review. *International Journal of Mechanical and Materials Engineering* 4(3): 239-248.

Akil, H. M., Omar, M. F., Mazuki, A. A. M., Safiee, S., Ishak, Z. A. M., and Abu Bakar, A. 2011. Kenaf fiber reinforced composites: A review. *Materials and Design* 32(8-9): 4107–4121.

Aldana, D. S., Villa, E. D., De Dios Hernández, M., Sánchez, G. G., Cruz, Q. R., Gallardo, S. F., Cruz, Q. R., Gallardo, S. F., Castillo, H. P., and Casarrubias, L. B. 2014. Barrier properties of polylactic acid in cellulose based packages using montmorillonite as filler. *Polymers 6*(9): 2386-2403.

Alemdar, A., and Sain, M. 2008. Biocomposites from wheat straw nanofibers: Morphology, thermal and mechanical properties. *Composites Science and Technology* 68(2): 557-565.

Ali, N. A., and Noori, F. T. M. 2014. Gas barrier properties of biodegradable polymer nanocomposites films. *Chemistry and Materials Research* 6(1): 44-51.

Ali, F., Chang, Y. W., Kang, S. C., and Yoon, J. Y. 2009. Thermal, mechanical and rheological properties of poly (lactic acid)/epoxidized soybean oil blends. *Polymer Bulletin 62*(1): 91-98.

Almenar, E., and Auras, R. 2010. Permeation, sorption, and diffusion in poly(lactic acid. In *Poly(lactic acid): Synthesis, Structures, Properties, Processing, and Applications (2nd ed.)*, eds. R. Auras, L. T. Lim, S. E. M. Selke, and H. Tsuji, pp. 155-179. Hoboken, New Jersey, US: John Wiley & Sons, Inc.

Al-Mulla, E. A. J., Ibrahim, N. A. B., Shameli, K., Ahmad, M. B., and Yunus, W. M. Z. W. 2014. Effect of epoxidized palm oil on the mechanical and morphological properties of a PLA–PCL blend. *Research on Chemical Intermediates* 40(2): 689-698.

Al-Mulla, E. A. J. 2011. Polylactic acid/epoxidized palm oil/fatty nitrogen compounds modified clay nanocomposites: Preparation and characterization. *Korean Journal of Chemical Engineering* 28(2): 620-626.

Al-Mulla, E. A. J., Yunus, W. M. Z. W., Ibrahim, N. A. B., and Rahman, M. Z. A. 2010. Properties of epoxidized palm oil plasticized polytlactic acid. *Journal of Materials Science* 45(7): 1942-1946.

Arbelaiz, A., Fernàndez, B., Ramos, J. A., and Mondragon, I. 2005. Thermal and crystallization studies of short flax fibre reinforced polypropylene matrix composites: Effect of treatments." *Thermochimica Acta* 440(2): 111-121.

Arrieta, M. P., López, J., Ferrándiz, S., and Peltzer, M. A. 2013. Characterization of PLA-limonene blends for food packaging applications. *Polymer Testing* 32(4): 760-768.

Arrieta, M. P., Samper, M. D., López, J., and Jiménez, A. 2014. Combined effect of poly (hydroxybutyrate) and plasticizers on polylactic acid properties for film intended for food packaging. *Journal of Polymers and the Environment* 22(4): 460-470.

Ashori, A., Harun., J., Raverty, W. D., and Mohd. Yusoff, M. N. 2006. Chemical and morphological characteristics of Malaysian cultivated kenaf (*Hibiscus cannabinus)* fiber. *Polymer-Plastics Technology and Engineering* 45(1): 131-134.

Atalla, R. H., and VanderHart, L. D. 1984. Native cellulose: A composite of two distinct crystalline forms. *Science* 223(4633): 283-285.

Auras, R., Harte, B., and Selke, S. 2004. An overview of polylactides as packaging materials. *Macromolecular Bioscience* 4(9): 835-864.

Ayranci, E., and Tunc, S. 2003. A method for the measurement of the oxygen permeability and the development of edible films to reduce the rate of oxidative reactions in fresh foods. *Food Chemistry* 80(3): 423-431.

Aziz, S. H., and Ansell, M. P. 2004. The effect of alkalization and fibre alignment on the mechanical and thermal properties of kenaf and hemp bast fibre composites: Part 1 - polyeste resin matrix. *Composites Science and Technology* 64(9): 1219-1230.

Bakis, C. E., Bank, L. C., Brown, V. L., Cosenza, E., Davalos, J. F., Lesko, J. J., Machida, A., Rizkalla, S. H., and Triantafillou, T. C. 2002. Fiber-reinforced polymer composites for construction-State-of-the-art review. *Journal of Composites for Construction* 6(2): 73-87.

Battegazzore, D., Bocchini, S., and Frache, A. 2011. Crystallization kinetics of poly(lactic acid)-talc composites. *EXPRESS Polymer Letters* 5(10): 849-858.

Bax, B., and Müssig, J. 2008. Impact and tensile properties of PLA/Cordenka and PLA/flax composites. *Composites Science and Technology* 68(7-8): 1601-1607.

Belmares, M., Blanco, M., Goddard, W. A., Ross, R. B., Caldwell, G., Chou, S.-H, Pham, J., Olofson, P. M, and Thomas, C. 2004. Hildebrand and Hansen solubility parameters from molecular dynamics with applications to electronic nose polymer sensors.*Journal of Computational Chemistry* 25(15): 1814-1826.

Beshay, A., and Hoa, S. V. 1992. Improved interface bonding between cellulosic fibers and thermoplastics. *Science and Engineering of Composite Materials* 2(2): 85-98.

Bibo, G. A., and Hogg, P. J. 1996. The role of reinforcement architecture on impact damage mechanisms and post-impact compression behaviour. *Journal of Materials Science* 31(5): 1115-1137.

Bigg, D. M. 1997. Effect of copolymer ratio on the crystallinity and properties of polylactic acid copolymers. In *Annual Technical Conference of the Society of Plastics Engineers*, 1996: 2028–2039.

Bisanda, E. T. N., and Ansell, M. P. 1991. The effect of silane treatment on the mechanical and physical properties of sisal-epoxy composites. *Composites Science and Technology* 41(2): 165-178.

BKH Consulting Engineers. *Towards the Establishment of a Priority List of Substances for Further Evaluation of Their Role in Endocrine Disruption (Final Report)*; European Commission DG Environment: Netherlands, 2006.

Bledzki, A. K., Mamun, A. A., Lucka-Gabor, M., and Gutowski, V. S. 2008. The effects of acetylation on properties of flax fibre and its polypropylene composites. *eXPRESS Polymer Letters* 2(6): 413–422.

Bledzki, A. K., and Gassan , J. 1999. Composites reinforced with cellulose based fibers. *Progress in Polymer Science* 24(2): 221-274.

Blum, F. D., Meesiri, W., Kang, H.-J., Gambogi, J. E. 1991. Hydrolysis, adsorption, and dynamics of silane coupling agents on silica sufaces. *Journal of Adhesion Science and Technology* 5(6): 479-496.

Borysiak, S., and Doczekalska, B. 2008. Research into the mercerization process of beechwood using the waxs method. *Fibres & Textiles in Eastern Europe* 16(71): 101-103.

Bourtoom, T. 2008. Plasticizer effect on the properties of biodegradable blend film from rice starch-chitosan. *Sonklanakarin Journal of Science and Technology* 30(1): 149-155.

Brochier Salon, M. C., Gerbaud, G., Abdelmouleh, M., Bruzzese, C., Boufi, S., and Belgacem, M. N. 2007. Studies of interactions between silane coupling agents and cellulose fibers with liquid and solid-state NMR. *Magnetic Resonance in Chemistry* 45(6): 473-483.

Brochier Salon, M. C., Abdelmouleh, M., Boufi, S., Belgacem, M. N., and Gandini, A. 2005. Silane adsorption onto cellulose fibers: Hydrolysis and condensation reactions. *Journal of Colloid and Interface Science* 289(1): 249-261.

Brosius, D. 2006. Natural fiber composites slowly take root. *Composites Technology*: 32-37.

Broström, J., Boss, A., and Chronakis, S. L. 2004. Biodegradable films of partly branched poly(l-lactide)-co-poly(epsilon-caprolactone) copolymer: Modulation of phase morphology, plasticization properties and thermal depolymerization. *Biomacromolecules* 5(3): 1124-1134.

Browning, B.L., 1967*. In Methods of Wood Chemistry (Vol. II)*. New York: Interscience Publishers.

Burgos, N., Martino, V. P., and Jiménez, A. 2013. Characterization and ageing study of poly(lactic acid) films plasticized with oligomeric lactic acid. *Polymer Degradation and Stability* 98(2): 651-658.

Butler, T. and Morris, B. A. 2013. PE based multilayer film Structures. In *Plastic Films in Food Packaging: Materials, Technology and Applications*, ed. S. Ebnesajjad, pp. 21-52. Oxford, UK: William Andrew Publishing.

Cao, X., Mohamed, A., Gordon, S. H., Willett, J. L., and Sessa, D. J. 2003. DSC study of biodegradable poly(lactic acid) and poly(hydroxy ester ether) blends. *Thermochimica Acta* 406(1-2): 115-127.

Cao, N., Yang, X., and Fu, Y. 2009. Effects of various plasticizers on mechanical and water vapor barrier properties of gelatin films. *Food Hydrocolloids 23*(3): 729-735.

Castellano, M., Gandini, A., Fabbri, P., and Belgacem, M. N. 2004. Modification of cellulose fibres with organosilanes: Under what conditions does coupling occur? *Journal of Colloid and Interface Science* 273(2): 505-511.

Chieng, B. W., Ibrahim, N. A., Wan Yunus, W. M. Z., Hussein, M. Z., and Silverajah, V. S. 2012. Graphene nanoplatelets as novel reinforcement filler in poly (lactic acid)/epoxidized palm oil green nanocomposites: Mechanical properties. *International Journal of Molecular Sciences* 13(9): 10920-10934.

Choudalakis, G., and Gotsis, A. D. 2009. Permeability of polymer/clay nanocomposites: A review. *European Polymer Journal* 45(4): 967-984.

Chua, S. C., Xu, X., and Guo, Z. 2012. Emerging sustainable technology for epoxidation directed toward plant oil-based plasticizers. *Process Biochemistry* 47(10): 1439-1451.

Commission of the European Communities, 1982. Council Directive of 18 October 1982 laying down the basic rules necessary for testing migration of the constituents of plastics materials and articles intended to come into contact with foodstuffs (82/711/EEC). *Official Journal of the European Communities*, L297: 26-30.

Commission of the European Communities, 1985. Council Directive of 19 December 1985 laying down the list of simulants to be used for testing migration of constituents of plastics materials and articles intended to come into contact with foodstuffs (85/572/EEC). *Official Journal of the European Communities*, L372: 14-21.

Courgneau, C., Domenek, S., Guinault, A., Avérous, L., and Ducruet, V. 2011. Analysis of the structure-properties relationships of different multiphase systems based on plasticized poly (lactic acid). *Journal of Polymers and the Environment* 19(2): 362-371.

Curie, K. J., Blemberg, R. J., Muehl, B. K., and Knoll, R. W. 2007. *U.S. Patent Application 11/769,777*.

Dai, X., Xiong, Z., Na, H., and Zhu, J. 2014. How does epoxidized soybean oil improve the toughness of microcrystalline cellulose filled polylactide acid composites? *Composites Science and Technology* 90: 9-15.

Daniel, J. R.1985. Cellulose, structure, and properties. In *Encyclopedia of Polymer Science and Engineering (Vol. 3)* pp. 90-123. New York, US: John Wiley & Sons, Inc.

Das, M. 2008. Effect of screw speed and plasticizer on the torque requirement in single screw extrusion of starch based plastics and their mechanical properties. *Indian Journal of Chemical Technology* 15(6): 555.

de Souza Lima, M. M., and Borsali, R. 2004. Rodlike cellulose microcrystals: Structure, properties, and applications. *Macromolecular Rapid Communications* 25(7): 771-787.

Di Lorenzo, M. L. 2006. Calorimetric analysis of the multiple melting behaviour of poly(L-lactic acid). *Journal of Applied Polymer Science* 100(4): 3145-3151.

Dinand, E., Vignon, M., Chanzy, H., and Heux, L. 2002. Mercerization of primary wall cellulose and its implication for the conversion of cellulose I→ cellulose II. *Cellulose* 9(1): 7-18.

Dittenber, D. B., and GangaRao, H. V. 2012. Critical review of recent publications on use of natural composites in infrastructure. *Composites Part A: Applied Science and Manufacturing* 43(8): 1419-1429.

Dufresne, A. 2008. Cellulose-based composites and nanocomposites. In *Monomers, Polymers and Composites from Renewable* Resources, eds. A. Gandini and M. N. Belgacem, pp. 401-418. Oxford, UK: Elsevier.

Dufresne, A. 2006. Comparing the mechanical properties of high performances polymer nanocomposites from biological sources. *Journal of Nanoscience and Nanotechnology* 6(2): 322-330.

Edeerozey, A. M., Akil, H. M., Azar, A. B., and Ariffin, M. I. Z. 2007. Chemical modification of kenaf fibers. *Materials Letters* 61(10): 2023-2025.

El-Shekeil, Y. A., Sapuan, S. M., Abdan, K., and Zainudin, E. S. 2012. Influence of fiber content on the mechanical and thermal properties of Kenaf fiber reinforced thermoplastic polyurethane composites. *Materials & Design* 40: 299-303.

Engel, L., Klingele, H., Ehrenstein, G. W., Schaper, H. 1981. In *An atlas of polymer damage: Surface examination by SEM*, pp. 136-211. London: Wolfe Publishing.

European Bioplastics, 2012. Market global production capacity of bioplastics. Retrieved 14 January 2013 from http://en.european-bioplastics.org/

European Commission, 2011. Commission Regulation (EU) No 10/2011 of 14 January 2011 on plastic materials and articles intended to come into contact with food. *Official Journal of European Community*, L12: 1–89.

European Union Reference Laboratory for Food Contact Materials (EURL-FCM) of Institute for Health and Consumer Protection (IHCP), European Commission. Test methods for overall migration into aqueous food simulants by article filling. Retrieved 26 Novemeber 2014 from http://ihcp.jrc.ec.europa.eu/our_labs/eurl_food_c_m/migration-methods

Fambri, L. and Migliaresi, C. 2010. Crystallization and thermal properties. In *Poly(lactic acid): Synthesis, Structures, Properties, Processing, and Applications (2nd ed.)*, eds. R. Auras, L. T. Lim, S. E. M.

Selke, and H. Tsuji, pp. 113-124. Hoboken, New Jersey, US: John Wiley & Sons, Inc.

Fang, J. M., Fowler, P. A., Escrig, C., Gonzalez, R., Costa, J. A., and Chamudis, L. 2005. Development of biodegradable laminate films derived from naturally occurring carbohydrate polymers. *Carbohydrate Polymers* 60(1): 39-42.

Fenollar, O., García, D., Sánchez, L., López, J., and Balart, R. 2009. Optimization of the curing conditions of PVC plastisols based on the use of an epoxidized fatty acid ester plasticizer. *European Polymer Journal* 45(9): 2674-2684.

Figura, L. O., and Teixeira, A. A. 2007. Permeability*. Food Physics (Physical Properties - Measurement and Applications),* pp. 233-255. Berlin, Germany: Springer-Verlag Berlin Heidelberg.

Fischer, E. W., Sterzel, H. J., and Wegner, G. 1973. Investigation of the structure of solution grown crystals of lactide copolymers by means of chemical reactions. *Kolloid-Zeitschrift und Zeitschrift für Polymere* 251(11): 980-990.

Formec International Ltd., n.d. Formec vacuum guide. Retrieved 10 January 2015 from http://inventionstudio.gatech.edu/wp/wp-content/uploads/2010/11/FormechVacuumGuide.pdf

Frigione, M. 2004. The transport of water and its effect on properties of cold-curing epoxy adhesives. In *Homolytic and Heterolytic Reactions: Problems and Solutions*, eds. E. Z. Gennadii, B. M. Iouorii, and J., Alfonso, pp. 283-305. New York, US: Nova Science Publishers, Inc.

Frone, A. N., Berlioz, S., Chailan, J. F., Panaitescu, D. M., and Donescu, D. 2011. Cellulose fiber-reinforced polylactic acid. *Polymer Composites* 32(6): 976-985.

Gaiolas, C., Costa, A. P., Nunes, M., Silva, M. J. S., and Belgacem, M. N. 2008. Grafting of Paper by Silane Coupling Agents Using Cold-Plasma Discharges. *Plasma Processes and Polymers* 5(5): 444-452.

Gamage, P. K., O'Brien, M., and Karunanayake, L. 2009. Epoxidation of some vegetable oils and their hydrolysed products with peroxyformic acid-optimised to industrial scale. *Journal of the National Science Foundation of Sri Lanka* 37(4): 229-240.

García, M. A., Martino, M. N., and Zaritzky, N. E. 2000. Lipid addition to improve barrier properties of edible starch-based films and coatings. *Journal of Food Science* 65(6): 941-944.

GB/T 5009.156-2003. *General principle for the determination of migration of packaging materials and their products*. National Standard of China.

Genomics, D. O. E. 2005. GTL Roadmap: Systems Biology for Energy and Environment. *US Department of Energy, Office of Science*. Retrieved on 15 June 2014 from http://genomicsgtl.energy.gov/roadmap/pdf/GenomicsGTL_Roadmap_highres.pdf

Georgopoulos, S. T., Tarantili, P. A., Avgerinos, E., Andreopoulos, A. G., and Koukios, E. G. 2005. Thermoplastic polymers reinforced with fibrous agricultural residues. *Polymer Degradation and Stability* 90(2): 303-312.

Geueke, B., Wagner, C. C., and Muncke, J. 2014. Food contact substances and chemicals of concern: a comparison of inventories. *Food Additives & Contaminants: Part A* 31(18): 1-13.

Ghosh, S. B., Bandyopadhyay-Ghosh, S., and Sai, M. 2010. Composites. In *Poly(lactic acid): Synthesis, Structures, Properties, Processing, and Applications (2nd ed.)*, eds. R. Auras, L. T. Lim, S. E. M. Selke, and H. Tsuji, pp. 293-310. Hoboken, New Jersey, US: John Wiley & Sons, Inc.

Global Packaging Alliance, n.d. Retrieved 14 January 2011 from http://www.global-packaging-alliance.com

Goda, K., Sreekala, M. S., Gomes, A., Kaji, T., and Ohgi, J. 2006. Improvement of plant based natural fibers for toughening green composites-Effect of load application during mercerization of ramie fibers. *Composites Part A: Applied Science and Manufacturin*g 37(12): 2213-2220.

Gon**ç**alves, C. M. B., Coutinho, J. A. P., and Marrucho, I. M. 2010. Optical properties. In *Poly(lactic acid): Synthesis, Structures, Properties, Processing, and Applications (2nd ed.)*, eds. R. Auras, L. T. Lim, S. E. M. Selke, and H. Tsuji, pp. 97-112. Hoboken, New Jersey, US: John Wiley & Sons, Inc.

Groot, W., van Krieken, J., Sliekersl, O., and de Vos, S. 2010. Production and purification of lactic acid and lactide. In *Poly(lactic acid): Synthesis, Structures, Properties, Processing, and Applications*

(2nd ed.), eds. R. Auras, L. T. Lim, S. E. M. Selke, and H. Tsuji, pp. 1-18. Hoboken, New Jersey, US: John Wiley & Sons, Inc.

Habibi, Y., El-Zawawy, W. K., Ibrahim, M. M., and Dufresne, A. 2008. Processing and characterization of reinforced polyethylene composites made with lignocellulosic fibers from Egyptian agro-industrial residues. *Composites Science and Technology* 68(7): 1877-1885.

HallStar, 2007. PLASTHALL® P-550 Plasticizer for Refrigerator Gaskets. Retrieved 28 April 2014 from http://www.hallstar.com/techdocs/plasthall_p550_for_refrigerator_gaskets.pdf

Han, S. O., Karevan, M., Sim, I. N., Bhuiyan, M. A., Jang, Y. H., Ghaffar, J., and Kalaitzidou, K. 2012. Understanding the reinforcing mechanisms in kenaf fiber/PLA and kenaf fiber/PP composites: A comparative study. *International Journal of Polymer Science* 2012: 1-8.

Hansen, C. M. 1967b. The three-dimensional solubility parameter-key to paint component affinities: solvents, plasticizers, polymers, and resins. II. Dyes, emulsifiers, mutual solubility and compatibility, and pigments. III. Independent calculation of the parameter components. *Journal of Paint Technology* 39(511): 505-510.

Hansen, C. M. 1967a. The three dimensional solubility parameter key to paint component affinities. I: Solvents, plasticisers, polymers and resins. *Journal of Paint Technology* 39(511): 104-117.

Hansen, C. M. 1969. The universality of the solubility parameter. *Industrial & Engineering Chemistry Product Research and Development* 8(1): 2-11.

Hansen, C. M., and Björkman, A. 1998. The ultrastructure of wood from a solubility parameter point of view. *Holzforschung-International Journal of the Biology, Chemistry, Physics and Technology of Wood* 52(4): 335-344.

Herrera-Franco, P., and Valadez-Gonzalez, A. 2005. A study of the mechanical properties of short natural-fiber reinforced composites. *Composites Part B: Engineering* 36(8): 597-608.

Hill, C. A., Khalil, H. P. S., and Hale, M. D. 1998. A study of the potential of acetylation to improve the properties of plant fibres. *Industrial Crops and Products* 8(1): 53-63.

Holbery, J., and Houston, D. 2006. Natural-fiber-reinforced polymer composites in automotive applications. *Journal of The Minerals, Metals & Materials Society* 58(11): 80-86.

Horvath, A. L. 2006. Solubility of structurally complicated materials: I. Wood. *Journal of Physical Chemistry* 35(1): 77-92.

Huda, M. S., Drzal, L. T., Mohanty, A. K., and Misra, M. 2007. Effect of fiber surface treatments on the properties of laminated biocomposites from poly(lactic acid) (PLA) and kenaf fibers. *Composites Science and Technology* 68(2): 424-432.

Ibrahim, N. A., Yunus, W. M. Z. W., Othman, M., Abdan, K., and Hadithon, K. A. 2009. “Poly(lactic acid)(PLA)-reinforced kenaf bast fibers composites: The effect of triacetin.” *Journal of Reinforced Plastics and Composites* 29(7): 1099-1111.

IS 9845:1998. *Determination of overall migration of constituents of plastics materials and articles intended to come in contact with foodstuffs - Method of analysis [PCD 12: Plastics]*. Bureau of Indian Standards. New Delhi, India.

Ismail, H., Shuhelmy, H., and Edyham, M. R. 2002. The effects of a silane coupling agent on curing characteristics and mechanical properties of bamboo fibre filled natural rubber composites. *European Polymer Journal* 38(1): 39-47.

ISO-11357-2. 2013. Plastics- Differential scanning calorimetry (DSC), Part 2: Determination of glass transition temperature and glass transition step height. International Organization for Standardization (ISO).

Jawaid, M. H. P. S., and Abdul Khalil, H. P. S. 2011. Cellulosic/synthetic fibre reinforced polymer hybrid composites: A review. *Carbohydrate Polymers* 86(1): 1-18.

Jensen, R. E., Palmese, G. R., and McKnight, S. H. 2006. Viscoelastic properties of alkoxy silane-epoxy interpenetrating networks. *International Journal of Adhesion and Adhesives* 26(1-2): 103-115.

Jia, P.-Y., Bo C.-Y., Hu, L-.H., and Zhou,Y.-H. 2014. Properties of poly(vinyl alcohol) plasticized by glycerin. *Journal of Forest Products and Industries* 3(3): 151-153.

Johansson, C., Bras, J., Mondragon, I., Nechita, P., Plackett, D., Simon, P., Svetec, D. G., Virtanen, S., Baschetti, M. G., Breen, C., Clegg, F., and Aucejo, S. 2012. Renewable fibers and bio-based materials for packaging applications - A review of recent developments. *BioResources* 7(2): 2506-2552.

John, M. J., and Thomas, S. 2008. Biofibres and biocomposites. *Carbohydrate Polymers* 71(3): 343-364.

Johnson, T. 2011. History of Composites: The Evolution of Lightweight Composite Materials. Retrieved 29 December 2011 from http://composite.about.com/od/aboutcompositesplastics/a/HistoryofComposites.htm

Joseph, P. V., Joseph, K., Thomas, S., Pillai, C. K. S., Prasad, V. S., Groeninckx, G., and Sarkissova, M. 2003. The thermal and crystallisation studies of short sisal fibre reinforced polypropylene composites. *Composites Part A: Applied Science and Manufacturing* 34(3): 253-266.

Joshi, S. V., Drzal, L. T., Mohanty, A. K., and Arora, S. 2004. Are natural fiber composites environmentally superior to glass fiber reinforced composites? *Composites Part A: Applied Science and Manufacturing* 35(3): 371-376.

Kalia, S., Kaith, B. S., and 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.

Kamper, S. L., and Fennema, O. 1984. Water vapor permeability of edible bilayer films. *Journal of Food Science* 49(6): 1478-1481.

Kargul, B., Çaglar, E., and Kabalay, U. 2004. Glass fiber-reinforced composite resin as fixed space maintainers in children: 12-month clinical follow-up. *Journal of dentistry for children (Chicago, Ill.)* 72(3): 109-112.

Karmakar, S. R. 1999. Chemical technology in the pre-treatment processes of textile. *Textile Science and Technology* 12: 279-315.

Kastinen, T. 2012. *Double Melting Behavior and Plasticization of Polylactic acid- A TOPEM and FTIR Study*, Master Thesis, Tampere University of Technology, Tampere.

Kazayawoko, M., Balatinecz, J. J., and Matuana, L. M. 1999. Surface modification and adhesion mechanisms in woodfiber-polypropylene composites. *Journal of Materials Science* 34(24): 6189-6199.

Khalid, M., Ratnam, C. T., Chuah, T. G., Ali, S., and Choong, T. S. 2008. Comparative study of polypropylene composites reinforced with oil palm empty fruit bunch fiber and oil palm derived cellulose. *Materials & Design* 29(1): 173-178.

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

Khan, M. A., and Hassan, M. M. 2006. Effect of γ-aminopropyl trimethoxy silane on the performance of jute–polycarbonate composites. *Journal of Applied Polymer Science* 100(5): 4142-4154.

Kim, H. S., Park, B. H., Choi, J. H., and Yoon, J. S. 2008. Mechanical properties and thermal stability of poly(L-lactide)/calcium carbonate composites *Journal of Applied Polymer* 109(5): 3087-3092.

Koenhen, D. M., and Smolders, C. A. 1975. The determination of solubility parameters of solvents and polymers by means of correlations with other physical quantities. *Journal of Applied Polymer Science* 19(4): 1163-1179.

Kofinas, P., Cohen, R. E., and Halasa, A. F. 1994. Gas permeability of polyethylene/poly(ethylene-propylene) semicrystalline diblock copolymers. *Polymer* 35(6): 1229–1235.

Koga, H., Kitaoka, T., and Isogai, A. 2011. In situ modification of cellulose paper with amino groups for catalytic applications *Journal of Materials Chemistry* 21(25): 9356-9361.

Krebs, J., Friedrich, K., and Bhattacharyya, D. 1998. A direct comparison of matched-die versus diaphragm forming. *Composites Part A: Applied Science and Manufacturing* 29(1): 183-188.

Kroon-Batenburg, L. M. J., and Kroon, J. 1997. The crystal and molecular structures of cellulose I and II. *Glycoconjugate Journal* 14(5): 677-690.

Kulinski, Z., and Piorkowska, E. 2005. Crystallization, structure and properties of plasticized poly (L-lactide). *Polymer* 46(23): 10290-10300.

Kulinski, Z., Piorkowska, E., Gadzinowska, K., and Stasiak, M. 2006. Plasticization of poly (L-lactide) with poly (propylene glycol). *Biomacromolecules* 7(7): 2128-2135.

Laboulfie, F., Hemati, M., Lamure, A., and Diguet, S. 2013. Effect of the plasticizer on permeability, mechanical resistance and thermal behaviour of composite coating films. *Powder Technology* 238: 14-19.

Langan, P., Nishiyama, Y., and Chanzy, H. 1999. A revised structure and hydrogen-bonding system in cellulose II from a neutron fiber diffraction analysis. *Jounal of the American Chemical Society* 121(43): 9940-9946.

Lathi, P. S., and Mattiasson, B. 2007. “Green approach for the preparation of biodegradable lubricant base stock from epoxidized vegetable oil. *Applied Catalysis B: Environmental* 69(3): 207-212.

Laxmeshwar, S. S., Viveka, S., Madhu Kumar, D. J., and Nagaraja, G. K. 2012. Preparation and properties of composite films from modified cellulose fiber-reinforced with PLA. *Pharma Chemica* 4(1, Cop): 159-168.

Leadbitter, J., Day, J. A., and Ryan, J. L. 1994. Compounding technology. In *PVC-Compounds, Processing and Applications. (Vol. 7, no. 6)*, RAPRA review reports: RAPRA Technology Limited, ed. R. Dolberry, pp. 26-32. Shropshire, UK: iSmithers

Lee, B.-H., Kim, H.-S., Lee, S., Kim, H.-J., and Dorgan, J. R. 2009. Bio-composites of kenaf fibers in polylactide: Role of improved I nterfacial adhesion in the carding process. *Composites Science and Technology* 69(15-16): 2573-2579.

Lee, S. N., Lee, M. Y., and Park, W. H. 2002. Thermal stabilization of poly (3-hydroxybutyrate) by poly (glycidyl methacrylate). *Journal of Applied Polymer Science* 83(13): 2945-2952.

Lee, S. Y., Kang, I. A., Doh, G. H., Yoon, H. G., Park, B. D., and Wu, Q. 2008. Thermal and mechanical properties of wood flour/talc-filled polylactic acid composites: Effect of filler content and coupling treatment. *Journal of Thermoplastic Composite Materials* 21(3): 209-223.

Liao, W. H., Hsiao, S. T., Wu, Y. F., Zeng, S. J., Li, S. M., Wang, Y. S., and Ma, C. C. M. (2014). Effect of silane-functionalized TiO 2 on the optical properties and moisture barrier of silicone resin nanocomposites. *RSC Advances* 4(73): 38614-38622.

Lim, L.-T., Auras, R., and Rubino, M. 2008. Processing technologies for poly(lactic acid). *Progress in Polymer Science* 33(8): 820-852.

Lim, L.-T., Cink, K., and Vanyo, T. 2010. Processing of (poly)lactic acid. In *Poly(lactic acid): Synthesis, Structures, Properties, Processing, and Applications (2nd ed.)*, eds. R. Auras, L. T. Lim, S. E. M. Selke, and H. Tsuji, pp. 191-216. Hoboken, New Jersey, US: John Wiley & Sons, Inc.

Lin, C. A., and Ku, T. H. 2008. Shear and elongational flow properties of thermoplastic polyvinyl alcohol melts with different plasticizer contents and degrees of polymerization. *Journal of Materials Processing Technology* 200(1): 331-338.

Ling, X., and Spruiell, J. E. 2006. Analysis of the complex thermal behavior of poly(L-lactic acid) film. I. Samples crystallized from the glassy state. *Journal of Polymer Science: Part B: Polymer Physics* 44(22): 3200-3214.

Liu, W., Mohanty, A. K., Drzal, L. T., Askel, P., and Misra, M. 2004. Effects of alkali treatment on the structure, morphology and thermal properties of native grass fibers as reinforcements for polymer matrix composites. *Journal of Materials Science* 39(3): 1051-1054.

Liu, L., Kerry, J. F., and Kerry, J. P. 2006. Effect of food ingredients and selected lipids on the physical properties of extruded edible films/casings. *International Journal of Food Science & Technology* 41(3): 295-302.

Liu, Y. P., and Hu, H. 2008. X-ray diffraction study of amboo fibers treated with NaOH.” *Fibers and Polymers* 9(6): 735-739.

Lu, J., Askeland, P., and Drzal, L. T. 2008. Surface modification of microfibrillated cellulose for epoxy composite applications. *Polymer* 49(5): 1285-1296.

Ludueña, L., Fasce, D., Alvarez, V. A., and Stefani, P. M. 2011. Nanocellulose from rice husk following alkaline treatment to remove silica. *BioResources* 6(2): 1440-1453.

Lung, C. Y. K., and Matinlinna, J. P. 2012. Aspects of silane coupling agents and surface conditioning in dentistry: An overview. *Dental Materials* 28(5): 467-477.

Luz, S. M., Del Tio, J., Rocha, G. J. M., Gonçalves, A. R., and Del’Arco

Jr., A. P. 2008. Cellulose and cellulignin from sugarcane bagasse reinforced polypropylene composites: Effect of acetylation on mechanical and thermal properties. *Composites: Part A* 39(9): 1362–1369.

Maizatul, N., Norazowa, I., Yunus, W. M. Z. W., Khalina, A., and Khalisanni, K. 2013. FTIR and TGA Analysis of Biodegradable Poly (Lactic Acid)/Treated Kenaf Bast Fibre: Effect of Plasticizers. *Pertanika Journal of Science & Technology* 21(1):151-160.

Majeed, K., Jawaid, M., Hassan, A., Bakar, A. A., Khalil, H. A., Salema, A. A., and Inuwa, I. 2013. Potential materials for food packaging from nanoclay/natural fibres filled hybrid composites. *Materials & Design* 46: 391-410.

Mansor, M. K., Ibrahim, N. A., Zin, W. M., Yunus, W. M. Z. W., and Ratnam, C. T. 2011. Effect of triacetin on the mechanical properties, morphology and water absorp-tion of poly(lactic acid)/tapioca starch composites. *Malaysian Polymer Journal* 6(2): 165-175.

Manthey, N. W., Cardona, F., Francucci, G., and Aravinthan, T. 2013. Thermo-mechanical properties of epoxidized hemp oil-based bioresins and biocomposites. *Journal of Reinforced Plastics and Composites* 32(19): 1444-1456.

Markarian, J. 2007. PVC additives- What lies ahead? *Plastics, Additives, and Compounding* 9(6): 22-25.

Martin, O., and Averous, L. 2001. Poly(lactic acid): plasticization and properties of biodegradable multiphase systems. *Polymer* 42(14): 6209-6219.

Martino, V. P., Jiménez, A., and Ruseckaite, R. A. 2009. Processing and characterization of poly(lactic acid) films plasticized with commercial adipates. *Journal of Applied Polymer Science* 112(4): 2010-2018.

Martin-Polo, M., Voilley, A., Blond, G., Colas, B., Mesnier, M., and Floquet, N. 1992. Hydrophobic films and their efficiency against moisture transfer. 2. Influence of the physical state. *Journal of Agricultural and Food Chemistry*40(3): 413-418.

Masirek, R., Kulinski, Z., Chionna, D., Piorkowska, E., and Pracella, M.

2007. Composites of poly(L-lactide) with hemp fibers: Morphology and thermal and mechanical properties. *Journal of Applied Polymer Science* 105(1): 255-268.

Mat Taib, R., Ramarad, S., Mohd Ishak, Z. A., and Todo, M. *Water Absorption and Tensile Properties of Kenaf Bast Fiber-Plasticized Poly(Lactic Acid) Biocomposites*. Paper presented at the Polymer Society 24th Annual Meeting (PPS-24), Salerno, Italy. 15-19 June, 2008.

Mathew, A. P., Oksman, K., and Sain, M. 2006. The effect of morphology and chemical characteristics of cellulose reinforcements on the crystallinity of polylactic acid. *Journal of Applied Polymer Science* 101(1): 300-310.

Mathlouthi, M. 2003. Packaging/packaging of solids. *In Encyclopedia of Food Sciences and Nutrition (2nd ed., Vol. 7)*, eds. B., Caballero, L. C., Trugo, and P. M., Finglas, pp. 4314. London, England: Academic Press.

McHugh, T. H., and Krochta, J. M. 1994. Dispersed phase particle size effects on water vapor permeability of edible emulsion films. *Journal of Food Processing and Preservation* 18(3): 173-188.

Meinander, K., Niemi, M., Hakola, J. S., and Selin, J. F. 1997. Polylactides-degradable polymers for fibres and films. *Macromolecular Symposia* 123(1): 147-153.

Mekonnen, T., Mussone, P., Khalil, H., and Bressler, D. 2013. Progress in bio-based plastics and plasticizing modifications*. Journal of Materials Chemistry A* 1(43): 13379-13398.

Meshram, P. D., Puri, R. G., and Patil, H. V. 2011. Epoxidation of wild safflower (Carthamus oxyacantha) oil with peroxy acid in presence of strongly acidic cation exchange Resin IR-122 as Catalyst. *International Journal of ChemTech Research* 3(3): 1152-1163.

Mohamad, M. 2010, February. Natural fibres for the 3rd millenium. *JEC Composites Magazine*, 24-28.

Mohanty, A. K., Misra, M., and Drzal, L. T. 2002. Sustainable bio-composites from renewable resources: opportunities and challenges in the green materials world. *Journal of Polymers and the Environment* 10(1-2): 19-26.

Mohanty, A. K., Misra, M., and Hinrichsen, G. 2000. Biofibres, biodegradable polymers and biocomposites: An overview. *Macromolecular Materials and Engineering* 276-277(1): 1-24.

Mohanty, A. K., Misra, M., Drzal, L. T., Selke, S. E., Harte, B. R., and Hinrichsen, G. 2005. Natural fibers, biopolymers and biocomposites: An Introduction. In *Natural Fibers, Biopolymers and Biocomposites*, eds. A. K. Mohanty, M. Misra and L. T. Drzal, pp. 1-36. London, UK: CRC Press.

Morye, S. S. 2005. A comparison of the thermoformability of a PPE/PP blend with thermoformable ABS. Part I: small deformation methods. *Polymer Engineering & Science* 45(10): 1369-1376.

Mrkić, S., Galić, K., Ivanković, M., Hamin, S., and Ciković, N. 2006. Gas transport and thermal characterization of mono-and di-polyethylene films used for food packaging. *Journal of Applied Polymer Science* 99(4): 1590-1599.

Munthoub, D. I., and Rahman, W. A. W. A. 2011. Tensile and water absorption properties of biodegradable composites derived from cassava skin/polyvinyl alcohol with glycerol as plasticizer. *Sains Malaysiana* 40(7): 713-718.

Murariu, M., Ferreira, A. D. S., Alexandre, M., and Dubois, P. 2008. Polylactide (PLA) designed with desired end-use properties: 1. PLA compositions with low molecular weight ester-like plasticizers and related performances. *Polymers for Advanced Technologies* 19(6): 636-646.

Mutasher, S. A., Poh, A., Than, A. M., and Law, J. 2011. The effect of alkali treatment mechanical properties of kenaf fiber epoxy composite. *Key Engineering Materials* 471-472: 191-196.

Mwaikambo, L. Y., and Ansell, M. P. 1999. The effect of chemical treatment on the properties of hemp, sisal, jute and kapok fibres for composite reinforcement. *Die angewandte makromolekulare Chemie* 272(1): 108-116.

Nakagaito, A. N., and Yano, H. 2005. Novel high-strength biocomposites based on microfibrillated cellulose having nano-order-unit web-like network structure. *Applied Physics A* 80(1): 155-159.

Nakagaito, A. N., Fujimura, A., Sakai, T., Hama, Y., and Yano, H. 2009. Production of microfibrillated cellulose (MFC)-reinforced polylactic acid (PLA) nanocomposites from sheets obtained by a papermaking-like process. *Composites Science and Technology* 69(7): 1293–1297.

NatureWorks LLC 2011. Processing guide of thermoforoming articles. Retrieved 15 December 2011 from http://www.natureworksllc.com/~/media/Technical_Resources/Processing_Guides/ProcessingGuide_Thermoforming-Articles_pdf.pdf

Nishida, H. 2010. Thermal degradation. In *Poly(lactic acid): Synthesis, Structures, Properties, Processing, and Applications (2nd ed.)*, eds. R. Auras, L. T. Lim, S. E. M. Selke, and H. Tsuji, pp. 401-412. Hoboken, New Jersey, US: John Wiley & Sons, Inc.

Nishino, T. 2004. Natural fibre sources. In *Green Composites: Polymer Composites and the Environment*, eds. C. Bailie, pp.49-80. Cambridge, UK: Woodhead Publishing Limited.

Oksman, K., Skrifvars, M., and Selin, J.-F. 2003. Natural fibres as reinforcement in polylactic acid (PLA) composites. *Composites Science and Technology* 63(9): 1317–1324.

Osterholtz, F. D., and Pohl, E. R. 1992. Kinetics of the hydrolysis and condensation of organofunctional alkoxysilanes: A review. *Journal of Adhesion Science and Technology* 6(1): 127-149.

Pamuła, E., Błażewicz, M., Paluszkiewicz, C., and Dobrzyński, P. 2001. FTIR study of degradation products of aliphatic polyesters–carbon fibres composites.*Journal of Molecular Structure* 596(1): 69-75.

Pan, P., Kai, W., Zhu, B., Dong, T., and Inoue, Y. 2007. Polymorphous crystallization and multiple melting behavior of poly(L-lactide): Molecular weight dependence. *Macromolecules* 40(19): 6898-6905.

Pang, A. L., and Ismail, H. 2013. Tensile properties, water uptake, and thermal properties of polypropylene/waste pulverized tire/kenaf (PP/WPT/KNF) composites. *BioResources* 8(1): 806-817

Pantoja, M., Díaz-Benito, B., Velasco, F., Abenojar, J., and del Real, J. C. 2009. Analysis of hydrolysis process of c-methacryloxypropyltrimethoxysilane and its influence on the formation of silane coatings on 6063 aluminum alloy. *Applied Surface Science* 255(12): 6386-6390.

Paszner, L. 1968. Effect of inter- and intra-crystalline swelling on cellulose degradation by gamma-rays. *Sven Papperstidn* 71(22): 822-828.

Pei, A., Zhou, Q., and Berglund, L. A. 2010. Functionalized cellulose nanocrystals as biobased nucleation agents in poly(L-lactide) (PLLA) - Crystallization and mechanical property effects. *Composites Science and Technology* 70(5): 815-821.

Petersen, K., Væggemose Nielsen, P., Bertelsen, G., Lawther, M., Olsen, M. B., Nilsson, N. H., and Mortensen, G. 1999. Potential of biobased materials for food packaging. *Trends in Food Science & Technology* 10(2): 52-68.

Petinakis, E., Simon, G., Dean, K., and Yu, L. 2013. Natural fibre bio-composites incorporating poly(lactic acid). *In Fiber Reinforced Polymers- The Technology Applied for Concrete Repair*, eds. M. Masuelli, pp. 41-59. Rijeka, Croatia: InTech Open Access Publisher.

Pillin, I., Montrelay, N., and Grohens, Y. 2006. Thermo-mechanical characterization of plasticized PLA: Is the miscibility the only significant factor? *Polymer* 47(13): 4676-4682.

Piorkowska, E., Kulinski, Z., Galeski, A., and Masirek, R. (2006). Plasticization of semicrystalline poly (L-lactide) with poly (propylene glycol). *Polymer*, *47*(20), 7178-7188.

Piszcek, K., 2005. Gelation of Suspension PVC with Small Amount of Plasticizer Compounds. *Polimery* 50(10): 765-768.

PLASTIVAL, *RTD-Based Cluster Initiative to Target Future Challenges of the Valencian Plastic Processing Industry;* CORDIS Project Reference 229935, European Commission: EU, 2009.

Plueddemann, E. P. 1988. Present status and research needs in silane coupling. In *Interfaces in Polymer, Ceramic, and Metal Matrix Composites*, ed. H. Ishida, pp. 17-33. New York: Elsevier.

Pluta, M. 2004. Morphology and properties of polylactide modified by thermal treatment, filling with layered silicates and plasticization. *Polymer* 45(24): 8239-8251.

Poly Print Inc. 2008. “Poly Print: Custom printers and laminators of flexible packaging. Understanding film properties. Retrieved 3 March 2013 from http://www.polyprint.com/

Pothan, L. A., Thomas, S., and Groeninckx, G. 2006. The role of fibre/matrix interactions on the dynamic mechanical properties of chemically modified banana fibre/polyester composites. *Composites: Part A* 37(9): 1260-1269.

Rahman, S. H. A., 2012. *Preparation and Characterization of Composite Films from Kenaf Derived Cellulose and Polylactic Acid (PLA),* Master Thesis, Universiti Putra Malaysia.

Rampinelli, G., Di Landro, L., and Fujii, T. 2010. Characterization of biomaterials based on microfibrillated cellulose with different modifications. *Journal of Reinforced Plastics and Composites* 29(12): 1793-1803.

Ranby, B. G. 1952. The physical characteristics of alpha-, beta- and gamma-cellulose. *Svensk Papperstid* 55(4): 115-124.

Rao, S., Jayaraman, K., and Bhattacharyya, D. 2011. Short fibre reinforced cores and their sandwich panels: Processing and evaluation. *Composites Part A: Applied Science and Manufacturing* 42(9): 1236-1246.

Ray, D., Sarkar, B. K., Rana, A. K., and Bose, N. R. 2001. The mechanical properties of vinylester resin matrix composites reinforced with alkali-treated jute fibres. *Composites Part A: Applied Science and Manufacturing* 32(1): 119-127.

Revol, J. F., and Goring, D. A. I. 1981. On the mechanism of the mercerization of cellulose in wood. *Journal of Applied Polymer Science* 26(4): 1275-1282.

Robertson, G. L. 2013. Packaging of dairy products. In *Food Packaging: Principles and Practice*, ed. G. L. Robertson, pp. 509-544. US: Taylor and Francis Group, LLC.

Rodriguez, E. S., Stefani, P., and Vazquez, A. 2007. Effects of fibers' alkali treatment on the resin transfer molding processing and mechanical properties of jute-vinylester composites. *Journal of Composite Materials* 41(14): 1729-1741.

Rong, M. Z., Zhang, M. Q., Liu, Y., Yang, G. C., and 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.

Rouison, D., Sain, M., and Couturier, M. 2004. Resin transfer molding of natural fiber reinforced composites: cure simulation. *Composites Science and Technology* 64(5): 629-644.

Rudnik, E. 2007. Thermal Properties of Biocomposites. *Journal of Thermal Analysis and Calorimetry* 88(2): 495-498.

Saad, M. J. 2011. Effect of maleated polypropylene (MAPP) on the tensile, impact and thickness swelling properties of kenaf core–polypropylene composites. *Journal of Science and Technology* 2(1): 33-44.

Salame, M., and Steingiser, S. 1977. Barrier polymers. *Polymer-Plastics Technology and Engineering* 8(2): 155–175.

Saremi, K., Tabarsa, T., Shakeri, A., and Babanalbandi, A. 2012. Epoxidation of soybean oil. *Annals of Biological Research* 3(9):4254-4258.

Satyanarayana, K. G., Sukumaran, K., Mukherjee, P. S., Pavithran, C., and Pillai, S. G. K. 1990. Natural fibre-polymer composites. *Cement and Concrete Composites* 12(2): 117-136.

Schirp, A., and Wolcott, M. P. 2005. Influence of fungal decay and moisture absorption on mechanical properties of extruded wood-plastic composites. *Wood and Fiber Science* 37(4): 643-652.

Sgriccia, N., Hawley, M. C., and Misra, M. 2008. Characterization of natural fiber surfaces and natural fiber composites. *Composites Part A: Applied Science and Manufacturing* 39(10): 1632-1637.

Shea, K. M. 2003. Pediatric exposure and potential toxicity of phthalate plasticizers. *Pediatrics* 111(6): 1467-1474.

Sheila, W. 2012. Association predicts fivefold growth of the bioplastics market by 2016. Retrieved 14 January 2013 from http://www.packaging-asia.com/article/association-predicts-fivefold-growth-of-the-bioplastics-market-by-2016/1380

Shellhammer, T. H., and Krochta, J. M. 1997. Whey protein emulsion film performance as affected by lipid type and amount. *Journal of Food Science* 62(2): 390-394.

Shen, C., Wang, Y., Li, M., and Hu, D. 2011. Crystal modifications and multiple melting behavior of poly (L-lactic acid-co-D-lactic acid). *Journal of Polymer Science Part B: Polymer Physics* 49(6): 409-413.

Shogren, R. 1997. Water vapor permeability of biodegradable polymers. *Journal of Environmental Polymer Degradation* 5(2): 91-95.

Silva, R. V., and Aquino, E. M. F. 2008. Curaua fiber: A new alternative to polymeric composites. *Journal of Reinforced Plastics and Composites* 27(1): 103-112.

Silverajah, V. S., Ibrahim, N. A., Zainuddin, N., Yunus, W. M. Z. W., and Hassan, H. A. 2012. Mechanical, thermal and morphological properties of poly (lactic acid)/epoxidized palm olein blend. *Molecules* 17(10): 11729-11747.

Silvestre, C., Duraccio, D., and Cimmino, S. 2011. Food packaging based on polymer nanomaterials. *Progress in Polymer Science* 36(12): 1766-1782.

Singh, B., Gupta, M., and Verma, A. 1996. Influence of fiber surface treatment on the properties of sisal-polyester composites. *Polymer Composites* 17(6): 910-918.

Siparsky, G. L., Voorhees, K. J., Dorgan, J. R., and Schilling, K. 1997. Water transport in polylactic acid (PLA), PLA/ polycaprolactone copolymers, and PLA/polyethylene glycol blends. *Journal of Environmental Polymer Degradation* 5(3): 125-136.

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

Siracusa, V. 2012. Food packaging permeability behaviour: A report. *International Journal of Polymer Science* 2012(1): 1-11.

Soulestin, J., Prashantha, K., Lacrampe, M. F., and Krawczak, P. 2011. Bioplastics based nanocomposites for packaging applications. In *Handbook of Bioplastics and Biocomposites Engineering Applications*, ed. S. Pilla, pp. 77-120. Salem, Massachusetts: Scrivener Publishing LLC.

Sreekala, M. S., Kumaran, M. G., and Thomas, S. 1997. Oil palm fibers: Morphology, chemical composition, surface modification, and mechanical properties. *Journal of Applied Polymer Science* 66: 821-835.

Sreekala, M. S., Kumaran, M. G., Joseph, S., Jacob, M., and Thomas, S. 2000. Oil palm fibre reinforced phenol formaldehyde composites: influence of fibre surface modifications on the mechanical performance. *Applied Composite Materials* 7(5-6): 295-329.

Srinivasa, P. C., Ramesh, M. N., and Tharanathan, R. N. 2007. Effect of plasticizers and fatty acids on mechanical and permeability characteristics of chitosan films. *Food Hydrocolloids* 21(7): 1113-1122.

Suardana, N. P. G., Piao, Y., and Lim, J. K. 2011. Mechanical properties of hemp fibers and hemp/PP composites: Effects of chemical surface treatment. *Materials Physics and Mechanics* 11(1): 1-8.

Sugiyama, J., Okano, T., Yamamoto, H., and Horii, F. 1990. Transformation of Valonia cellulose crystals by an alkaline hydrothermal treatment. *Macromolecules* 23(12): 3196-3198.

Tábi, T., Sajó, I. E., Szabó, F., Luyt, A. S., and Kovács, J. G. 2010. Crystalline structure of annealed polylactic acid and its relation to processing. *eXPREss Polymer Letters* 4(10): 659-668.

Talib, R. A., Tawakkal, I. S. M. A., and Abdan, K. 2011. The influence of mercerised kenaf fibres reinforced polylactic acid composites on dynamic mechanical analysis. *Key Engineering Materials* 471-472: 815-820.

Tang, B*. Fiber Reinforced Polymer Composites Applications in USA*. Paper presented at the First Korea/ U.S.A. Road Workshop Proceedings, Washington, D.C. 28-29 January, 1997.

TAPPI, 1999. *Alpha-, Beta- and Gamma-Cellulose in Pulp*; Technical Association of the Pulp and Paper Industry.

Tawakkal, I. S. M., Talib, R. A., Abdan, K., and Chin, N. L. 2012. Mechanical and physical properties of kenaf-derived cellulose (KDC)-filled polylactic acid (PLA) composites. *BioResources* 7(2): 1643-1655.

Tawakkal, I. S. M. A., Talib, R. A., Khalina, A., Chin, N. L., and Ibrahim, M. N. 2010. Optimisation of processing variables of kenaf derived cellulose reinforced polylactic acid. *Asian Journal of Chemistry* 22(9): 6652-6662.

Tee, Y. B., Talib, R. A., Abdan, K., Chin, N. L., Basha, R. K., and Yunos, K. F. M. 2013. Thermally grafting aminosilane onto kenaf-derived cellulose and its influence on the thermal properties of poly (lactic acid) composites. *BioResources* 8(3): 4468-4483.

Téllez, G. L., Vigueras-Santiago, E., and Hernández-López, S. 2009. Characterization of linseed oil epoxidized at different percentages. *Superficies Y Vacío* 22(1): 5-10.

Thomason, J. L., and Vlug, M. A. 1996. Influence of fibre length and concentration on the properties of glass fibre-reinforced polypropylene: 1. Tensile and flexural modulus. C*omposites Part A: Applied Science and Manufacturing* 27(6): 477-484.

Timothy, V. D. 2011. Applications of nanotechnology in food packaging and food safety: Barrier materials, antimicrobials, and sensors. *Journal of Colloid and Interface Science* 363(1): 1-24.

Tokoro, R., Vu, D. M., Okubo, K., Tanaka, T., Fujii, T., and Fujiura, T. 2008. How to improve mechanical properties of polylactic acid with bamboo fibers. *Journal of Materials Science* 43(2): 775-787.

Urayama, H., Moon, S.-I., Kimura, Y. 2003. Microstructure and thermal properties of polylactides with different L- and D-unit sequences: Importance of the helical nature of the L-sequenced segments. *Macromolecular Materials and Engineering* 288(2): 137-143.

Valadez-Gonzalez, A., Cervantes-Uc, J. M., Olayo, R., and Herrera-Franco, P. J. 1999. Chemical modification of henequen fibers with an organosilane coupling agent. *Composites Part B: Engineering* 30(3): 321-331.

van Tuil, R., Fowler, P., Lawther, M., and Weber, C. J. 2000. Properties of biobased packaging materials. In .) *Biobased Packaging Material for the Food Industry- Status and Perspectives*, ed. C. J. Weber, pp. 13-44. Frederiksberg, Denmark: KVL.

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

Wang, Q., Chen, Y., Tang, J., and Zhang, Z. 2013. Determination of the solubility parameter of epoxidized soybean oil by inverse gas chromatography. *Journal of Macromolecular Science, Part B* 52(10): 1405-1413.

Wang, Y., Qi, R., Xiong, C., and Huang, M. 2011. Effects of coupling agent and interfacial modifiers on mechanical properties of poly (lactic acid) and wood flour biocomposites. *Iranian Polymer Journal* 20(4): 281-294.

Wang, Y., Tong, B., Hou, S., Li, M., amd Shen, C. 2011. Transcrystallization behavior at the poly (lactic acid)/sisal fibre biocomposite interface. *Composites Part A: Applied Science and Manufacturing* 42(1): 66-74.

Wang, Z. G., Hsiao, B. S., Sauer, B. B., and Kampert, W. G. 1999. The nature of secondary crystallization in poly (ethylene terephthalate). *Polymer* 40(16): 4615-4627.

Wang, Z., Hou, X., Mao, Z., Ye, R., Mo, Y., and Finlow, D. E. 2008. Synthesis and characterization of biodegradable poly (lactic acid-co-glycine) via direct melt copolymerization. *Iranian Polymer Journal* 17(10): 791-798.

Wasanasuk, K., and Tashiro, K. 2011. Crystal structure and disorder in Poly (l-lactic acid) δ form (α′ form) and the phase transition mechanism to the ordered α form. *Polymer* 52(26): 6097-6109.

Weigel, C., and Kellner, R. 1989. FTIR-ATR-spectroscopic investigation of the silanization of germanium surfaces with 3-aminopropyltriethoxysilane.*Fresenius' Zeitschrift für analytische Chemie* 335(7): 663-668.

Wells, J. K., and Beaumont, P. W. 1985. Crack-tip energy absorption processes in fibre composites. *Journal of Materials Science* 20(8): 2735-2749.

Westman, M. P., Fifield, L. S., Simmons, K. L., Laddha, S. G., and Kafentzis, T. A. 2010. Natural fiber composites: A review. (No. 19220) Springfield, U.S.A.: Pacific Northwest National Laboratory.

Wilson, K., Ringstrom, E., and Hedlund, I. 1955. The alkali solubility of pulp. *Svensk Papperstid* 55(2): 31-37.

Wojciechowska, E., Fabia, J., Slusarczyk, C., Gawlowski, A., Wysocki, W. and Graczyk, T., 2005. Processing and supermolecular structure of new iPP/PLA fibres. *FIBRES & TEXTILES in Eastern Europe* 13(5): 126-128.

Wong, S., Shanks, R. A., and Hodzic, A. 2003. Poly (L-lactic acid) composites with flax fibers modified by plasticizer absorption. *Polymer Engineering & Science* 43(9): 1566-1575.

Woo, E. M., Sun, Y. S., and Yang, C. P. 2001. Polymorphism, thermal behavior, and crystal stability in syndiotactic polystyrene vs. its miscible blends. *Progress in Polymer Science* 26(6): 945-983.

Wötzel, K., Wirth, R., and Flake, M. 1999. Life cycle studies on hemp fibre reinforced components and ABS for automotive parts. *Die Angewandte Makromolekulare Chemie* 272(1): 121-127.

Xanthos, M. 1983. Processing conditions and coupling agent effects in polypropylene/wood flour composites. *Plastics and Rubber Processing and Applications* 3(3): 223-228.

Xie, Y., Hill, C. A., Xiao, Z., Militz, H., and Mai, C. 2010. Silane coupling agents used for natural fiber/polymer composites: A review. *Composites Part A: Applied Science and Manufacturing* 41(7): 806-819.

Xiong, Z., Li, C., Ma, S., Feng, J., Yang, Y., Zhang, R., and Zhu, J. 2013c. The properties of poly (lactic acid)/starch blends with a functionalized plant oil: Tung oil anhydride. *Carbohydrate Polymers* 95(1): 77-84.

Xiong, Z., Ma, S., Fan, L., Tang, Z., Zhang, R., Na, H., and Zhu, J. 2014. Surface hydrophobic modification of starch with bio-based epoxy resins to fabricate high-performance polylactide composite materials. *Composites Science and Technology* 94, 16-22.

Xiong, Z., Yang, Y., Feng, J., Zhang, X., Zhang, C., Tang, Z., and Zhu, J. 2013a. Preparation and characterization of poly (lactic acid)/starch composites toughened with epoxidized soybean oil. *Carbohydrate Polymers* 92(1): 810-816.

Xiong, Z., Zhang, L., Ma, S., Yang, Y., Zhang, C., Tang, Z., and Zhu, J. 2013b. Effect of castor oil enrichment layer produced by reaction on the properties of PLA/HDI-g-starch blends. *Carbohydrate Polymers* 94(1): 235-243.

Xu, Y. Q., and Qu, J. P. 2009. Mechanical and rheological properties of epoxidized soybean oil plasticized poly (lactic acid). *Journal of Applied Polymer Science* 112(6): 3185-3191.

Yamamoto, Y., Inoue, Y., Onai, T., Doshu, C., Takahashi, H., and Uehara, H. 2007. Deconvolution analyses of differential scanning calorimetry profiles of β-crystallized polypropylenes with synchronized X-ray measurements. *Macromolecules* 40(8): 2745-2750.

Yan, S., Yin, J., Yang, Y., Dai, Z., Ma, J., and Chen, X. 2007. Surface-grafted silica linked with l-lactic acid oligomer: a novel nanofiller to improve the performance of biodegradable poly (l-lactide). *Polymer* 48(6): 1688-1694.

Yang, S. L., Wu, Z. H., Meng, B., and Yang, W. 2009. The effects of dioctyl phthalate plasticization on the morphology and thermal, mechanical, and rheological properties of chemical crosslinked polylactide. *Journal of Polymer Science Part B: Polymer Physics* 47(12): 1136-1145.

Yew, G. H., Yusof, A. M., Ishak, Z. M., and Ishiaku, U. S. 2005. Water absorption and enzymatic degradation of poly (lactic acid)/rice starch composites. *Polymer Degradation and Stability* 90(3): 488-500.

Yoon, J.-S., Jung, H.-W., Kim, M.-N., and Park, E.-S. 2000. Diffusion coefficient and equilibrium solubility of water molecules in biodegradable polymers. *Journal of Applied Polymer Science* 77(8): 1716-1722.

Younes, H., and Cohn, D. 1988. Phase separation in poly (ethylene glycol)/poly (lactic acid) blends. *European Polymer Journal* 24(8): 765-773.

Zakaria, Z., Islam, M. S., Hassan, A., Mohamad Haafiz, M. K., Arjmandi, R., Inuwa, I. M., and Hasan, M. 2013. Mechanical properties and morphological characterization of PLA/chitosan/epoxidized natural rubber composites. *Advances in Materials Science and Engineering* 2013: 1-7.

Zhang, S. M., Liu, J., Zhou, W., Cheng, L., and Guo, X. D. 2005. Interfacial fabrication and property of hydroxyapatite/polylactide resorbable bone fixation composites. *Current Applied Physics* 5(5): 516-518.

Zhao, Y., Qiu, J., Feng, H., and Zhang, M. 2012. The interfacial modification of rice straw fiber reinforced poly (butylene succinate) composites: Effect of aminosilane with different alkoxy groups. *Journal of Applied Polymer Science* 125(4): 3211-3220