UNIVERSITI PUTRA MALAYSIA

PREPARATION AND CHARACTERIZATION OF BIOCOMPOSITES FROM OIL PALM MESOCARP FIBER AND POLY(BUTYLENE SUCCINATE)

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FS 2015 44

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Doctor of Philosophy
Universiti Putra Malaysia
2015

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By

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

August 2015

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

PREPARATION AND CHARACTERIZATION OF BIOCOMPOSITES FROM OIL PALM MESOCARP FIBER AND POLY(BUTYLENE SUCCINATE)

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Chairman: Associate Professor Nor Azowa Ibrahim, PhD
Faculty : Science

In the present work, biocomposites were prepared from poly(butylene succinate) (PBS) and various weight percentages (10-70 wt%) of oil palm mesocarp fiber (OPMF) by using a melt blending technique followed by hot-press moulding. Surface modification of OPMF was carried out via four approaches, namely alkali, superheated steam, combination superheated steam-alkali, and bleaching treatments, aiming at improving the interfacial adhesion between the hydrophilic OPMF and the hydrophobic PBS. Additionally, silane coupling agent of 3-aminopropyltrimethoxysilane (APS) was also introduced into the biocomposite system to induce some chemical linkage between OPMF and PBS. Apart from that, electron beam irradiation (EBI) was also applied to improve the tensile properties of OPMF/PBS biocomposites.

The results indicated that the tensile, flexural, and impact properties of OPMF/PBS biocomposites were comparable and even better than those of OPMF/polypropylene (OPMF/PP) and oil palm empty fruit bunch fiber/PBS (OPEFBF/PBS) biocomposites. Additionally, the biodegradation rate of OPMF/PBS biocomposites was also noticeably higher than those of OPMF/PP biocomposites, and showed comparable values to those of OPEFBF/PBS biocomposites.

The chemical compositions of OPMF were changed after various treatment processes as validated by chemical analysis and Fourier transform infrared (FTIR) spectroscopy. The treated OPMF under microscopy observation showed relatively rough texture surfaces due to the elimination of impurities and hemicellulose. The crystallinity index and thermal stability of treated OPMF were relatively higher than that of untreated OPMF as determined by using X-ray diffraction (XRD) analysis and thermogravimetric analysis (TGA), respectively. A reduction in water uptake of fiber after treatments was also noted. The OPMF treated at 5 wt% NaOH solution for 180 min, and superheated steam temperature of 220 °C for 60 min gave biocomposites with best combinations of tensile strength, tensile modulus, and elongation at break. The subsequent alkali treatment of superheated steam-treated OPMF with 2 wt% NaOH for

180 min enhanced further the tensile properties of the corresponding biocomposite. The interfacial adhesion between treated OPMF and PBS was improved considerably as indicated by scanning electron micrographs. The treated OPMF/PBS biocomposites also exhibited higher thermal and dimensional stabilities, as well as more resistance to microorganism attacks in comparison to that of untreated OPMF/PBS biocomposite.

The addition of 2 wt% APS into the PBS biocomposite filled with combination superheated steam-alkali treated OPMF further enhanced the tensile, flexural, and impact strengths by 16, 26, and 8%, respectively, relatively to biocomposite without APS addition. Similarly, the resistance to water uptake and thickness swelling of this biocomposite was improved by 34 and 49%, respectively. The SEM observation of the tensile fractured surface showed that APS further improved the interfacial adhesion between combination superheated steam-alkali-treated OPMF and PBS. Some chemical linkages have been formed between the treated OPMF, PBS, and APS, mainly via hydrogen bonding as indicated by FTIR spectroscopy.

The biocomposites fabricated from PBS and bleached OPMF showed improvement of 54, 830, and 43% in tensile strength, tensile modulus, and elongation at break as compared to that of untreated OPMF. In addition, bleached OPMF/PBS biocomposites also showed better flexural and impact performance in comparison to that of untreated OPMF/PBS biocomposite.

For EBI treatment, the results indicated that OPMF/PBS biocomposites irradiated with 20 kGy of applied dosage showed a considerable improvement of 47% in tensile strength, 772% in tensile modulus and 28% in elongation at break relative to non-irradiated OPMF/PBS biocomposite. The water uptake and thickness swelling of OPMF/PBS biocomposites were also reduced after EBI treatment.

The OPMF/PBS biocomposites fabricated from this work showed potential application in particleboards, and dashboard for car interior compartment.

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

PENYEDIAAN DAN PENCIRIAN BIOKOMPOSIT DARIPADA SERAT SABUT KELAPA SAWIT DAN POLI(BUTILENA SUKSINAT)

Oleh

THEN YOON YEE

Ogos 2015

Pengerusi : Profesor Madya Nor Azowa Ibrahim, PhD
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Dalam kajian ini, biokomposit telah disediakan daripada poli(butilena suksinat) (PBS) dan pelbagai peratusan berat (10-70 wt%) serat sabut kelapa sawit (OPMF) dengan menggunakan teknik pengadunan leburan diikuti oleh acuan panas-tekan. Pengubahsuaian permukaan OPMF telah dilaksanakan melalui empat pendekatan, iaitu rawatan alkali, stim panas lampau, gabungan stim panas lampau-alkali, dan pelunturan, dengan tujuan untuk meningkatkan lekatan-antara-muka di antara hidrofilik OPMF dan hidrofobik PBS. Selain itu, agen gandingan silana 3-aminopropiltrimetosisilana (APS) juga dimasukkan ke dalam sistem biokomposit untuk mendorong pembentukan ikatan kimia antara OPMF dan PBS. Tambahan pula, sinaran alur elektron (EBI) juga digunakan untuk memperbaiki sifat ketegangan biokomposit OPMF/PBS.

Keputusan menunjukkan bahawa sifat-sifat rintangan ketegangan, kelenturan, dan hentaman bagi biokomposit OPMF/PBS adalah setanding malah lebih baik daripada biokomposit serat sabut kelapa sawit/polipropilena (OPMF/PP) dan serat tandan kosong kelapa sawit/PBS (OPEFB/PBS). Tambahan pula, kadar biodegradasi bagi biokomposit OPMF/PBS juga nyata lebih tinggi berbanding dengan biokomposit OPMF/PP, dan setanding dengan biokomposit OPEFBF/PBS.

Komposisi kimia OPMF telah berubah selepas pelbagai proses rawatan seperti disahkan oleh analisis kimia dan spektroskopi jelmaan inframerah Fourier (FTIR). Di bawah pemerhatian mikroskop, OPMF terawat menunjukkan permukaan tekstur kasar disebabkan penyingkiran benda asing dan hemiselulosa. Indeks penghabluran dan kestabilan terma OPMF terawat secara relatif lebih tinggi daripada OPMF tanpa terawat seperti yang ditentukan dengan menggunakan analisis pembelauan sinar-X (XRD) dan analisis termogravimetri (TGA), masing-masing. Selepas rawatan, pengurangan pengambilan air bagi serat juga tercatat. Rawatan OPMF dengan menggunakan 5 wt% larutan NaOH selama 180 minit, dan suhu stim panas lampau 220 °C selama 60 minit, memberikan biokomposit dengan kombinasi terbaik dari segi kekuatan ketegangan, modulus ketegangan, dan pemanjangan pada saat putus.

Rawatan alkali seterusnya pada OPMF stim panas lampau-terawat dengan menggunakan 2 wt% larutan NaOH selama 180 min, meningkatkan lagi sifat-sifat ketegangan biokomposit tersebut. Lekatan-antara-muka di antara OPMF terawat dan PBS telah bertambah baik dengan ketara seperti yang ditunjukkan dalam mikrograf imbasan elektron. Biokomposit OPMF terawat/PBS juga mempamerkan kestabilan terma dan dimensi yang lebih tinggi, serta lebih tahan terhadap serangan mikroorganisma berbanding dengan biokomposit OPMF tanpa terawat/PBS.

Penambahan 2 wt% APS ke dalam biokomposit PBS yang terisi dengan OPMF gabungan stim panas lampau-alkali terawat mempertingkatkan lagi rintangan ketegangan, kelenturan, hentaman sebanyak 16, 26, dan 8%, masing-masing, berbanding dengan biokomposit tanpa APS. Begitu juga, biokomposit ini juga menunjukkan peningkatan sebanyak 34 dan 49% terhadap rintangan untuk pengambilan air dan pembengkakan ketebalan. Pemerhatian pada permukaan putus ketegangan dengan SEM menunjukkan kehadiran APS meningkatkan lagi lekatan-antara-muka di antara OPMF gabungan stim panas lampau-alkali terawat dan PBS. Spektroskopi FTIR menunjukkan ikatan kimia terbentuk di antara OPMF terawat, PBS dan APS, terutamanya melalui ikatan hidrogen.

Biokomposit yang dibuat daripada PBS dan OPMF terluntur menunjukkan peningkatan sebanyak 54, 830, dan 43% dalam kekuatan ketegangan, modulus ketegangan, dan pemanjangan pada saat putus berbanding dengan OPMF tanpa terawat. Di samping itu, biokomposit OPMF terluntur/PBS juga menunjukkan rintangan kelenturan dan hentaman yang lebih baik berbanding dengan biokomposit OPMF tanpa terawat/PBS.

Bagi rawatan EBI, keputusan menunjukkan bahawa biokomposit OPMF/PBS yang tersinar dengan dos 20 kGy menunjukkan peningkatan yang besar sebanyak 47% dalam kekuatan ketegangan, 772% dalam modulus ketegangan dan 28% dalam pemanjangan pada saat putus berbanding dengan biokomposit tanpa sinaran. Pengambilan air dan pembengkakan ketebalan biokomposit juga telah dikurangkan selepas rawatan EBI.

Biokomposit OPMF/PBS yang disediakan dalam kajian ini menunjukkan potensi aplikasi dalam papan partikel dan papan pemukaan untuk ruang dalaman kereta.

ACKNOWLEDGEMENTS

First and foremost, I would like to express my sincere gratitude and appreciation to my supervisor Associate Professor Dr. Nor Azowa Ibrahim for her valuable guidance, suggestions, assistance, patience and encouragement for the past four years. Her dedication and enthusiasm in science greatly encourage me to work hard and pursue a great goal in research.

I am grateful to my co-supervisors Dr. Norhazlin Zainuddin, Dr. Hidayah Ariffin, and Prof. Dato' Dr. Wan Md Zin Wan Yunus as their fruitful and infectious enthusiasm certainly filled me with positive energy whenever I had discussions with them.

I would like to express my deepest gratitude to my beloved family especially my parents who have always believes in me, and endured with me during difficult times.

I would like to acknowledge the Ministry of Education, Malaysia, for the provision of a MyPhD scholarship, under MYBRAIN 15 from September 2011 to February 2015.

My sincere appreciation is extended to the helpful and encouraging laboratory officers in Departments of Chemistry as well as Physic, Faculty of Science, Universiti Putra Malaysia especially Madam Rusnani Amirudin, Madam Zaidina Mohd Daud, Madam Nor Azlina Shari, Madam Noriza Maslan and Mr. Nordin Abdul Kadir for sharing their time and willingness to help in analysis of samples. I extend my appreciation to all staffs in the Electron Microscopy Unit, Institute of Bioscience, Universiti Putra Malaysia, especially Madam Zahidah Muhamed for assisting in the scanning electron microscopy analysis.

I am also grateful to all my friends and colleagues in polymer research group especially Dr. Lim Chee Siong, Dr. Chieng Buong Woei, Dr. Phang Pei See, Mr. Chee Wei Kit, Mr. Eng Chern Chiet, Mr. Teh Cher Chean, Mr. Lim Chaw Jiang, Mr. Lai Fok How, Miss Teh Siew Nu, Miss Liau Cha Ping, Miss Marwah Rayung and Miss Nur Inani Abdul Razak who have always giving help, suggestions, encouragement and companion during hard times of my study. Million thanks also extended to Madam Noor Ida Amalina Ahamad Nordin and Mr. Mohd Nor Faiz Norahim from Faculty of Biotechnology and Biomolecular Science, Universiti Putra Malaysia for assisting in fiber collection and superheated steam treatment.

Acknowledgements also given to Dr. Khairul Zaman Hj Mohd Dahlan and Mr. Mohd Faiza Abdul Rahman from Malaysian Institute for Nuclear Technology Research for providing the dashboard panels and assisting in electron beam irradiation treatment.

Last but not the least, million thanks as well for those are involved directly or indirectly in the process of completing this thesis.

I certify that a Thesis Examination Committee has met on 21 August 2015 to conduct the final examination of Then Yoon Yee on his thesis entitled "Preparation and Characterization of Biocomposites from Oil Palm Mesocarp Fiber and Poly(butylene succinate)" 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 degree of Doctor of Philosophy.

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TABLE OF CONTENTS

4 INFLUENCE OF FIBER CONTENT ON PROPERTIES OF OIL PALM MESOCARP FIBER/POLY(BUTYLENE SUCCINATE) BIOCOMPOSITES

5 A COMPARATIVE STUDY ON PROPERTIES OF OIL PALM MESOCARP FIBER/ POLY(BUTYLENE SUCCINATE), OIL PALM MESOCARP FIBER/POLYPROPYLENE, AND OIL PALM EMPTY FRUIT BUNCH FIBER/ POLY(BUTYLENE SUCCINATE) BIOCOMPOSITES

6 INFLUENCE OF FIBER TREATMENT PARAMETERS ON TENSILE PROPERTIES AND DIMENSIONAL STABILITY OF OIL PALM MESOCARP FIBER/POLY(BUTYLENE SUCCINATE) BIOCOMPOSITES

7 PROPERTIES OF SUPERHEATED STEAM-, ALKALI-, AND SUPERHEATED STEAM-ALKALI-TREATED OIL PALM MESOCARP FIBER: A COMPARATIVE SUDY

8 A COMPARATIVE STUDY ON PROPERTIES OF SUPERHEATED STEAM-, ALKALI-, AND SUPERHEATED STEAM-ALKALI-TREATED OIL PALM MESOCARP FIBER/ POLY(BUTYLENE SUCCINATE) BIOCOMPOSITES

9 INFLUENCE OF 3-AMINOPROPYLTRIMETHOXYSILANE ON PROPERTIES OF SUPERHEATED STEAM-ALKALI-TREATED OIL PALM MESOCARP FIBER/POLY(BUTYLENE SUCCINATE) BIOCOMPOSITES

LIST OF TABLES

LIST OF FIGURES

LIST OF ABBREVIATIONS

APS	3-aminopropyltrimethoxysilane
ASTM	American Standards for Testing Methods
ATR	Attenuated total reflection
BMF	Bleached OPMF
CrI	Crystallinity index
DMA	Dynamic mechanical analysis
DSC	Differential scanning calorimetry
DTG	Differential thermogravimetric
EBI	Electron beam irradiation
FFB	Fresh fruit bunch
FTIR	Fourier transform infrared
GMA	Glycidyl methacrylate
GMA-g-PBS	Glycidyl methacrylate-grafted-PBS
HDPE	High density polyethylene
LDPE	Low density polyethylene
MA	Maleic anhydride
MA-g-PBS	Maleic anhydride-grafted-PBS
NMF	NaOH-treated OPMF
OPEFBF	Oil palm empty fruit bunch fiber
OPKS	Oil palm kernel shell
OPLF	Oil palm leaf fiber
OPMF	Oil palm mesocarp fiber
PBAT	Poly(butylene adipate-co-terephthalate)
PBS	Poly(butylene succinate)
PCL	Polycaprolactone
PE	Polyethylene
PHB	Polyhydroxybutyrate

PHBV	Polyhydroxybutyrate-co-valerate
phr	Parts to hundred parts rubber
PLA	Polylactic acid
POME	Palm oil mill effluent
PP	Polypropylene
PVP	Poly(vinylpyrrolidone)
SEM	Scanning electron microscopy
SMF	Superheated steam-treated OPMF
SHS	Superheated steam
SNMF	Superheated steam-NaOH-treated OPMF
TGA	Thermogravimetric analysis
UMF	Untreated OPMF
XRD	X-ray diffraction

CHAPTER 1

INTRODUCTION

1.1 Background of Study

The majorities of plastic products used today are made from non-renewable petrochemical resources and are not biodegradable. These products can contribute to environmental pollution long after their functional life, particular to littering and waste disposal problems. This has triggered a search for biodegradable plastics, especially those derived from renewable resources, as substitutes for petrochemical-based as well as non-biodegradable plastics (Song *et al.*, 2009).

In recent years, several types of biodegradable plastics have been introduced into the market. However, due to their relatively high production costs, these types of plastics cannot economically compete with the conventional plastics already present in the market (Singh *et al.*, 2008).

Several attempts have been made by researchers to combine these biodegradable plastics with inexpensive fillers, particularly natural fibers obtained from agricultural wastes, to more cost-effectively produced biocomposites without changing their biodegradable characteristic (Ibrahim *et al.*, 2009; Hamid *et al.*, 2010; Rayung *et al.*, 2014; Razak *et al.*, 2014). This type of biocomposites can provide a feasible solution to recently growing environmental threats as well as a sustainable solution to replace plastics or composite materials those made from limited petrochemical resources.

As compared to synthetic fibers, natural fibers possess several advantages, which include low cost and low density, biodegradability, eco-friendliness, renewability, and high specific strength and stiffness relative to synthetic fibers (Wambua *et al.*, 2003; Terzopoulou *et al.*, 2014). Additionally, natural fibers can be obtained at a relatively low energy input with a lesser amount of CO_2 emissions; they cause little abrasion to machinery during processing compared to synthetic fibers (Samir *et al.*, 2005).

According to recent publications, a wide range of natural fibers including fibers from kenaf (Ibrahim *et al.*, 2010; Thirmizir *et al.*, 2011; Lee *et al.*, 2013; Sis *et al.*, 2013; Razak *et al.*, 2014), oil palm empty fruit bunch (Ibrahim *et al.*, 2009; Hamid *et al.*, 2010; Rayung *et al.*, 2014), rice straw (Wu and Liao, 2012), coir (Nam *et al.*, 2011), flax (Bax and Mussig, 2008; Barkoula *et al.*, 2010), coconut (Wu, 2010), jute (Liu *et al.*, 2009a; Nam *et al.*, 2012), hemp (Sawpan *et al.*, 2011; Terzopoulou *et al.*, 2014), and bamboo (Singh *et al.*, 2008) have been compounded with various types of biodegradable plastics, particularly polylactic acid (PLA), poly(butylene succinate) (PBS), polycaprolactone (PCL), polyhydroxybutyrate (PHB), and polyhydroxybutyrate-co-valerate (PHBV) to fabricate low-cost and biodegradable biocomposites.

1.2 Statement of Problem

Malaysia is known as one of the largest palm oil producers and exporters in the world. In the palm oil industry, many oil palm biomasses are produced and left either in the plantation area or palm oil mills after extraction of palm oil. These oil palm biomasses include various fibrous materials from frond, trunk, empty fruit bunch, mesocarp, and leaf. Amongst them, oil palm mesocarp fiber (OPMF) is of interest here. It was reported that the amount of OPMF generated in Malaysia in 2014 was 13.1 million ton. Currently, OPMF is used as a low cost boiler fuel to produce steam in generating electricity for self-supplying the palm oil mills and is under-utilized commercially. However, due to the incomplete combustion, dark smoke is produced while burning OPMF and give rise to the environmental problems. In this work, the OPMF will be utilized in a more sustainable way, by compounding it with thermoplastic to fabricate biocomposites. This could be a key role to reduce the environmental problems caused by burning OPMF while developing a value added product. At the moment, very little research has been conducted on OPMF, especially with respect to the fabrication of biocomposites. Thus, there is no doubt that the use of OPMF as filler to develop biocomposites will be an added value of OPMF in the future.

Natural fibers are relatively heat sensitive and its degradation initiates at temperature of approximately 160 °C. At the moment, the biodegradable plastics of PLA, PHB, and PHVB have relatively high processing temperature ranging from 150 to 180 °C, thus degradation of fiber might occur during melt processing and impart adverse effect on the properties of the biocomposites. Although PCL has relatively lower melting temperature of 60 °C, but this property has limited applications due to its softening effect at elevated temperature. In this work, the thermoplastic of PBS was chosen as a matrix due to its biodegradability and relatively lower melting temperature than those of PLA, PHB, and PHBV, at which degradation to fiber could be minimized during the compounding process. Additionally, PBS is also commercially available at prices lower than those of PLA, PHB, and PHBV. These advantages make it an attractive alternative compared to those of other biodegradable and non-biodegradable thermoplastics.

Though many works have been carried out on natural fiber-filled biodegradable plastic biocomposites, however these biocomposites were mainly fabricated using natural fiber content less than or equal to that of 50 wt%. Thus these biocomposites may not be cost effective as the price of the biodegradable plastics is relatively higher than that of natural fiber. In this study, biocomposites were first fabricated from PBS and various weight percentages of OPMF (10 to 70 wt%) via a melt blending method followed by hot-press moulding. Later, biocomposite with OPMF-to-PBS at weight ratio of 70:30 was used for further study in this work, aiming at producing the biocomposite materials at a competitive price.

Biocomposites made from biodegradable plastics and natural fibers are fully biodegradable and price-competitive, yet this combination has several limitations, including poor interfacial adhesion between the two phases due to their dissimilar surface properties, and poor dimensional stability resulting from the high moisture uptake of fiber itself. This often leads to biocomposites with relatively poor mechanical properties and dimensional stability. It is known that the primary attraction of biocomposites market nowadays is the competitive price of natural fibers. Therefore, low-cost and efficient modification processes for natural fibers are of importance in producing strong materials at a competitive price. In this work, alkali (NaOH) and superheated steam (SHS) treatments were chosen to modify the surface of fiber because these treatments appear to be relatively simple and inexpensive but effective to modify the surface of fiber at relatively low environmental impact. To date, there is no study on the utilization of NaOH- and SHS-treated OPMF in biocomposite fabrication has been reported. Additionally, the OPMF was also treated by using combination SHS-NaOH treatments, aiming at reducing the NaOH consumption. A silane coupling agent of 3-aminopropyltrimethoxysilane (APS) was also introduced into the biocomposite system to induce some chemical linkage between OPMF and PBS. In this work, bleaching treatment was also carried out on OPMF, aiming at increasing the brightness of OPMF. Apart from that, electron beam irradiation was also used as a post treatment process to improve the tensile properties of OPMF/PBS biocomposites.

1.3 Significance of Study

This thesis is very practical as it concerns on producing environmentally friendly biocomposite materials from biodegradable thermoplastic and natural fiber via a melt mixing technique followed by hot-press molding. The selection of OPMF in this study is a good effort in order to reduce a bulk of mesocarp fiber waste in Malaysia. Momentarily, very little research has been conducted on OPMF, especially with respect to the fabrication of biocomposites. Most of the studies on OPMF were focused on biosugar production. Thus, there is no doubt that the use of OPMF as filler to develop biocomposites will be an added value of OPMF in the future.

1.4 Objectives of Study

The objectives of the present works are as follow:

1. To prepare biocomposites from PBS and OPMF at various weight percentages, as well as to characterize their dimensional stability, mechanical, thermal, and morphological properties.
2. To determine the potential of OPMF and PBS as biocomposite materials in comparison to those of oil palm empty fruit bunch fiber/PBS (OPEFBF/PBS) and OPMF/polypropylene (OPMF/PP) biocomposites in terms of mechanical properties, dimensional stability, and biodegradability.
3. To modify the OPMF surface via superheated steam, alkali, and combination superheated steam-alkali treatments, as well as to characterize their physical, chemical, thermal, and morphological properties after each treatment processes.
4. To prepare biocomposites from superheated steam-, alkali-, and superheated steam-alkali-treated OPMF and PBS, and characterize their dimensional stability, mechanical, thermal, and morphological properties.
5. To evaluate the effect of 3-aminopropyltrimethoxysilane on dimensional stability, mechanical, thermal, and morphological properties of chemically treated OPMF/PBS biocomposite.

6. To examine the influence of bleaching treatment on properties of OPMF and its effect on the biocomposites dimensional stability, mechanical, thermal, and morphological properties.
7. To determine the effect of electron beam irradiation treatment on dimensional stability, tensile, and morphological properties of OPMF/PBS biocomposite.

1.5 Organization of Thesis

Chapter 1 introduces the background, statement of problem, scope of study, significance of study, and objectives of this research. Chapter 2 gives the literature review related to this research. Chapter 3 describes the details on the materials and methods use to carry out this research. Chapter 4 discusses influence of fiber content on physical, mechanical, thermal, and morphological properties of OPMF/PBS biocomposites (Paper VIII). Chapter 5 highlights comparative study between OPMF/PBS, OPMF/PP, and OPEFBF/PBS biocomposites in terms of their mechanical properties, dimensional stability, and biodegradability (Paper I). Chapter 6 describes influence of OPMF treatment parameters on biocomposite's tensile properties and dimensional stability. Chapter 7 concerns the effect of various surface treatments of OPMF on its morphology, chemical composition, crystallinity, thermal stability, and water absorption behaviors (Paper III). Chapter 8 reports the comparative study of treated OPMF/PBS biocomposites in terms of physical, mechanical, thermal, and morphological properties (Paper II, V, and VI). Chapter 9 discusses effect of silane coupling agent on physical, mechanical, thermal, and morphological properties of treated OPMF/PBS biocomposite (Paper IX). Chapter 10 describes the ability of bleaching process of OPMF at improving the biocomposite's physical appearance, mechanical properties as well as dimensional stability (Paper VII). Chapter 11 concerns effect of electron beam irradiation on tensile properties and dimensional stability of OPMF/PBS biocomposite (Paper IV). Chapter 12 describes briefly the potential application of the biocomposites prepared from this work. Chapter 13 gives the conclusions of this research and recommendation for future work.

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