Faculty of Engineering

PREPARATION AND CHARACTERIZATION OF SAGO STARCH-CELLULOSE REINFORCED POLYLACTIC ACID HYBRID BIOCOMPOSITE

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PREPARATION AND CHARACTERIZATION OF SAGO STARCH-CELLULOSE REINFORCED POLYLACTIC ACID HYBRID BIOCOMPOSITE

MUHAMMAD JAYYAD

A dissertation submitted in partial fulfilment
of the requirement for the degree of
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Dedicated to my beloved parents and my family who always bestow me sustainable motivations and encouragements

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ABSTRACT

Plastics are one of the most heavily used materials that are used in everyday life and have numerous applications. The majority of plastics are petroleum-based which poses a problem in their disposal due to the fact that they are non-biodegradable. A new type of material that has shown great promise as an alternative is bioplastics (also called biopolymers or biocomposites). Biocomposites are based on renewable sources, biodegradable and exhibit superior mechanical, morphological and chemical properties. They also have vast applications including packaging and biomedical applications. Polylactic Acid (PLA) is one of the leading biopolymer that is used in biocomposites as matrix phase whereas sago starch and cellulose are among the most promising biopolymers to be used as the filler/reinforcement phase. This research attempted to synthesize and characterize a hybrid biocomposite by using PLA as the matrix phase and the sago starch and cellulose as the filler phase. Furthermore, the characterization of the hybrid biocomposite was done by carrying out the Tensile Testing, FTIR Analysis, SEM Analysis, DSC Analysis, TGA Analysis and Water Absorption Test. The mechanical, thermal, morphological and structural properties as well as water absorption resistance of the hybrid biocomposite have been tested and analyzed for four different sago starch-cellulose hybrid filler loads including 10wt%, 20wt%, 30wt% and 35wt% both with and without chemical modifications. The optimum hybrid filler loading for yielding the best tensile, morphological, structural and thermal properties and moisture absorption resistance of the hybrid biocomposite was found to be 35wt% sago starch-cellulose loading. The chemical modification using 1,2-Epoxy-5-Hexene of the sago starch-cellulose hybrid biofillers into the PLA matrix significantly improved the hybrid biocomposite and were strongly considered to be potentially utilized in a variety of applications including biomedical applications, tissue engineering, food packaging applications, used as plastic bags and disposable cutlery as well as in making baby nappies.

Keywords: Biocomposite, Sago Starch, Cellulose, Polylactic Acid, 1,2-Epoxy-5-Hexene, Filler, Matrix

ABSTRAK

Plastik adalah salah satu bahan yang paling banyak digunakan yang digunakan dalam kehidupan sehari-hari dan mempunyai pelbagai aplikasi. Majoriti plastik yang berasaskan petroleum yang menimbulkan masalah dalam pelupusan mereka disebabkan oleh hakikat bahawa mereka tidak terbiodegradasi. Sejenis baru bahan yang telah menunjukkan harapan besar sebagai alternatif adalah bioplastik (juga dipanggil biopolimer atau biocomposites). Biocomposites adalah berdasarkan kepada sumber-sumber yang boleh diperbaharui, boleh dilupuskan dan mempamerkan sifat-sifat mekanikal, morfologi dan kimia yang unggul. Mereka juga mempunyai aplikasi yang luas termasuk pembungkusan dan aplikasi bioperubatan. asid polylactic (PLA) adalah salah satu biopolimer utama yang digunakan dalam biocomposites sebagai fasa matriks manakala kanji sagu dan selulosa adalah antara biopolimer paling menjanjikan untuk digunakan sebagai fasa pengisi / pengukuhan. Kajian ini cuba untuk mensintesis dan mencirikan biokomposit hibrid dengan menggunakan PLA sebagai fasa matriks dan kanji sagu dan selulosa sebagai fasa pengisi. Tambahan pula, pencirian biokomposit hibrid itu dilakukan dengan menjalankan ujian tegangan, Analisis FTIR, Analisis SEM, Analisis DSC, Analisis TGA dan Ujian Penyerapan Air. Sifat-sifat mekanik, haba, morfologi dan struktur serta rintangan penyerapan air biokomposit hibrid telah diuji dan dianalisis untuk empat muatan sagu kanji selulosa hibrid pengisi yang berbeza termasuk 10wt%, 20wt%, 30wt% dan 35wt% kedua-duanya dengan dan tanpa pengubahsuaian kimia. Hibrid optimum pengisi loading untuk menghasilkan tegangan yang terbaik, morfologi, ciri-ciri struktur dan terma dan rintangan penyerapan kelembapan biokomposit hibrid didapati 35wt% sagu kanji selulosa loading. Pengubahsuaian kimia menggunakan 1,2-Epoxy-5-Hexene daripada sagu biofillers kanji selulosa hibrid ke dalam matriks PLA yang bertambah baik dengan ketara biokomposit hibrid dan amat dianggap sebagai berpotensi digunakan dalam pelbagai aplikasi termasuk aplikasi bioperubatan, kejuruteraan tisu, aplikasi pembungkusan makanan, menggunakan beg plastik dan kutleri pakai buang serta dalam membuat lampin bayi.

Kata Kunci: Biokomposit, Sago kanji, selulosa, asid polylactic, 1,2-Epoxy-5-Hexene, Pengisi, Matrix

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ABBREVIATIONS

DSC	:	Differential Scanning Calorimetry
TPS	:	Thermoplastic Starch
FTIR	:	Fourier Transform Infrared
TGA	:	Thermogravimetric Analysis
MAPP	:	Maleated Polypropylene
NaOH	:	Sodium Hydroxide
SiH_4	:	Silicon Hydride
PLA	:	Poly(lactic) Acid
PCL	:	Polycaprolactone
rpm	:	Revolution per Minute
SEM	:	Scanning Electron Microscopy
PMC	:	Polymeric Matrix Material
MMC	:	Metal Matrix Material
CMM	:	Ceramic Matrix Material
FRC	:	Fiber Reinforced Composite
DCM	:	Dicholoromethane
PVOH		Polyvinyl Alcohol
PET		Polyethylene Tetraphthalate
DP		Degree of Polymerization

NOMENCLATURE

%	:	Percent
°C	:	Degree Celsius
cm^{-1}	:	Per centimetre
m	:	Metres
g	:	Gram
GPa	:	Giga Pascals
mg	:	Milligrams
kN	:	Kilo Newton
L	:	Litre
mL	:	Millilitre
MPa	:	Mega Pascal
nm	:	Nanometer
rpm	:	Revolution per Minute
wt%	:	Weight Percentage
μm	:	Micrometer
g/mol	:	Grams per Moles
ºC/min	:	Degree Celsius per Minute
mm/min	:	Millimetres per Minute
m^2	:	Metres Squared

s	:	Seconds
h	:	Hours
Deg	:	Degree
N	:	Newton

CHAPTER 1

INTRODUCTION

1.1 Thesis Overview

This project/thesis is divided into five chapters. Chapter one covers the background of the study, the problem statement, the research objectives, the scope of the study, expected outcomes from this research and a Gantt chart to keep track of the research project progress. The Gannt chart is provided in **Appendix A**. The second chapter is on the literature review which covers the recent developments in the field of biocomposites using various type fillers and matrix phases, and includes the classifications and properties of the sago starch, cellulose and PLA respectively. The third chapter covers the methodology of the experimental works including the preparation and characterization of the hybrid biocomposite that is constructed for this research project. The fourth chapter covers the results obtained from carrying out the experiments and discusses them in detail. The fifth chapter presents recommendations and conclusion of the project.

1.2 Background of Study

Plastic is a typical organic polymer that is derived from petrochemicals, which are produced and used globally. This is mainly because of their low energy consumption during fabrication, their inertness and their ease of processing, which in turn makes them highly suitable to be used extensively in all areas (Sarifuddin et al., 2013). Despite that, the rapid expansion and fast growing production in plastic material consumption have resulted in major problems in the plastic disposal management. It is known that plastic is inert to the microorganism or the chemicals in the environment and thus, the disposal of very large amounts of plastic possess huge and critical environmental problems since plastic takes a significantly long period of time to decompose. Concerns about global environmental issues have caused a necessity to substitute the petroleum-based materials by the sustainable renewable resources in order to minimize the negative ecological and environmental impacts of polymeric materials (Abdulkhani et al., 2015). In recent years, particular attention has been given for the development of biodegradable polymers from the renewable resources especially for the purpose of disposable and packaging applications in order to maintain the sustainable development of ecologically and economically attractive technology and also towards a greener surrounding environment (Zuraida et al., 2012). Furthermore, the environmental problems worldwide and the rapid depletion of hydrocarbons resources, which are utilized in the manufacturing of petroleum-derived plastics, are insisting on a much more sustainable development of green composites (also called environmental friendly materials). Therefore, in tackling the environmental issues related to the conventional petrochemical-based plastic and its disposal, steps has been taken in recent years to develop biodegradable materials (Sarifuddin et al., 2013). Biodegradable materials can be synthetic, natural or combination of both.

For several years, the development of innovative and novel biopolymers has been carried out and continues to be an area of interest for many modern scientists and researchers (Hoque et al., 2013). In general, green composites comprise of biodegradable plant derived polymers that are used as matrixes and biodegradable plant derived fibres that are used as fillers (Wattnzkornsiri & Tongnunui, 2013). A composite is considered to be biodegradable if these integral parts are biodegradable. One of the most promising alternatives to the conventional petrochemical-based plastics is the Polylactic Acid (PLA). PLA is essentially a renewable natural resource based biodegradable thermoplastic polymer that is environmental friendly, biocompatible, sustainable and has wide range of medical, drug delivery, and textile or packaging applications which makes it attractive to researchers and industry especially in the food packaging and biomedical sectors (Nuthong et al., 2013). It is actually a type of commercial synthetic biopolymer made from L- and D-lactic acids, which are derived from the fermentation of carbohydrate crops such as corn starch (Qu et al., 2010). PLA also has good mechanical properties, thermal plasticity, good biocompatibility and processability, high strength and modulus and is readily fabricated, making it a promising polymer for various end-use applications (Sumigin et al., 2012). Also, PLA is one of the main representatives of the biodegradable polymers due to its good mechanical and optical properties (Hajba et al., 2015). Many researchers have found cellulose as one of the best choice of biodegradable reinforcement for PLA composites due to the fact that it is a tough, fibrous and water-insoluble material (Murphy & Collins, 2016). Cellulose is a natural polymer that is obtained from a renewable resource and is one of the most abundant organic compounds found in nature (Oka, 2010). Besides cellulose, another natural polymer that is very promising and has attracted the attention of researchers in the field of material science is starch. Starch is a polysaccharide polymer of D-anhydroglucose repeating units containing two main constituents, i.e. amylose and amylopectin (Wattanakornsiri & Tongnunui, 2013). Sago starch is an attractive source and a promising raw material for the development of PLA composites since it is naturally renewable, cheap, and abundant (Wattanakornsiri & Tongnunui, 2013).

Thus, this research was carried out to synthesis and characterize the sago starch-cellulose reinforced PLA hybrid biocomposite by using sago starch and cellulose as the filler phases and PLA as the matrix phase and to analyse the mechanical, physical, chemical, morphological and structural properties of the hybrid biocomposite. This research also focused and covered on the chemical modifications of the filler phase of the hybrid biocomposite. Comparisons between the chemically modified and unmodified hybrid biocomposites were made as well.

1.3 Problem Statement

Despite the advantages of PLA, it has deficiencies which include brittleness, low impact strength, poor thermal stability and low ability in resisting thermal deformation. To improve these properties, the focus has been shifted towards employing biodegradable reinforcements into the PLA matrix to produce a fully biocompatible biocomposite material or green composite with enhanced physical, mechanical, chemical and morphological properties. PLA is widely being used as a matrix phase in a biocomposite whereas natural polymers are being used as the reinforced or filler phase. Over the years, many efforts have been exerted to develop