

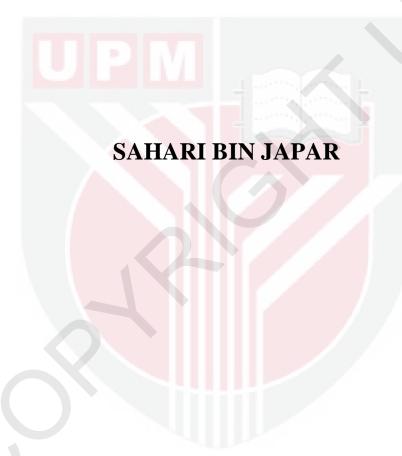
UNIVERSITI PUTRA MALAYSIA

CHARACTERIZATION AND DEVELOPMENT OF BIOPOLYMER DERIVED FROM ARENGA PINNATA AND THEIR BIOCOMPOSITES

SAHARI BIN JAPAR

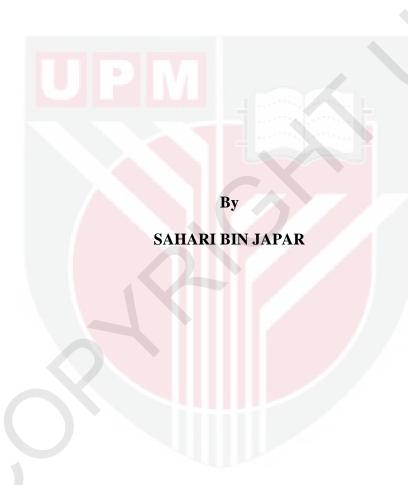
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DOCTOR OF PHILOSOPHY UNIVERSITI PUTRA MALAYSIA

CHARACTERIZATION AND DEVELOPMENT OF BIOPOLYMER DERIVED FROM ARENGA PINNATA AND THEIR BIOCOMPOSITES



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

DEDICATIONS

For all your advice and encouragement, this thesis is gratefully dedicated to my beloved parent, family and my friends. Thank you very much for your continuous support and effort towards the publication of this thesis.



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

CHARACTERIZATION AND DEVELOPMENT OF BIOPOLYMER DERIVED FROM ARENGA PINNATA AND THEIR BIOCOMPOSITES

By

SAHARI BIN JAPAR

May 2013

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This research was carried out to produce a new biopolymer (plasticized SPS) and biocomposites (SPF/SPS) derived from sugar palm tree. The plasticized SPS and SPF/SPS biocomposites were produced by a compression moulding method. Due to unique behavior of starch, the important properties of SPS have been studied in order to explore their potential to be used as a new alternative biopolymer. It was observed that the starches isolated from sugar palm tree contain comparable amounts of amylose (37.60%) and thermally stable than other starches. Then, SPS was added with 15 to 40 w/w% of glycerol to prepare workable bioplastics. Generally, the addition of glycerol decreases the transition temperature of plasticized SPS and the mechanical properties (i.e. tensile, flexural and impact) of plasticized SPS increase with the increasing of glycerol but up to 30 w/w%. Meanwhile, the water absorption of plasticized SPS decreases with increasing of glycerol. The effect of fiber content (i.e. 10%, 20% and 30% by weight percent) on mechanical properties, water absorption behavior and thermal properties of SPF/SPS biocomposites were investigated. The mechanical properties of plasticized SPS improved with the incorporation of fibers. Fiber loading also increased the thermal stability of the biocomposite in this investigation. Water uptake and moisture content of SPF/SPS biocomposites decreased with the incorporation of fibers, which is due to better

interfacial bonding between the matrix and fibers as well as the hindrance to absorption caused by the fibers. Morphological studies through scanning electron microscopy showed homogeneous distribution of fibres and matrix with good adhesion which play an important role in improving the mechanical properties of biocomposites. But the mechanical properties of the biocomposite dropped after being stored for 72 hours at 75% relative humidity and large cracks appear on the surface of the matrix (SPS) due to water absorption. Finally, biodegradable and weathering testing were also studied for both biopolymer and biocomposite. The environmental effect on plasticized SPS and SPF/SPS biocomposite were lost 78.09% and 53.67% tensile strength respectively at the end of 72 h of the weathering testing period. The biodegradation test shows that the plasticized SPS degrades very quickly and losses 63.58% weight at the end of 72 h compared to SPF/SPS biocomposites.

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

PENCIRIAN DAN PEMBANGUNAN BIOPOLIMER YANG DIPEROLEH DARIPADA ARENGA PINNATA DAN BIOKOMPOSITNYA

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Penyelidikan ini dijalankan untuk menghasilkan biopolimer (SPS terplastik) dan biokomposit (SPF/SPS) baru yang diterbitkan daripada pokok enau. SPS terplastik dan SPF/SPS biokomposit ini dihasilkan dengan menggunakan kaedah pengacuan mampatan. Disebabkan sifat kanji yang unik, sifat-sifat kanji yang penting dikaji untuk meneroka potensinya sebagai alternatif biopolimer yang baru. Hasil penyelidikan menunjukkan bahawa kanji daipada pokok enau mengandungi amilosa yang setanding dan sifat haba yang stabil berbanding kanji-kanji yang diperolehi daripada sumber yang lain. Kemudian, SPS ditambah dengan gliserol sebanyak 15 hingga 40% mengikut berat untuk menghasilkan bioplastik yang boleh digunakan. Secara keseluruhannya, penambahan gliserol menguranggkan suhu peralihan SPS terplastik dan sifat-sifak mekanikal (tegangan, lenturan dan impak) SPS terplastik bertambah dengan pertambahan gliserol sehinggal 30% mengikut berat. Manakala penyerapan air SPS terplastik berkurang dengan pertambahan gliserol. Kesan kandungan gentian (iaitu 10%, 20% and 30% mengikut berat) terhadap sifat-sifat mekanikal, penyerapan air dan haba bagi SPF/SPS biokomposit juga dikaji. Sifatsifat mekanikal SPS terplastik bertambah baik dengan penambahan gentian. Penambahan gentian juga meningkatkan kestabilan haba dalam kajian ini.

Penyerapan air dan kandungan lembapan SPF/SPS biokomposit menurun dengan penambahan gentian yang disebabkan oleh ikatan yang lebih baik antara matriks dan gentian serta sifat menghalang penyerapan yang disebabkan oleh gentian. Kajian morfologi melalui imbasan mikroskop elektron menunjukkan taburan seragam gentian dan matriks dengan ikatan yang baik yang memainkan peranan penting dalam meningkatkan sifat-sifat mekanikal biokomposit. Tetapi sifat-sifat mekanikal biokomposit menurun selepas disimpan selama 72 jam pada kelembapan relatif 75% dan retakan yang besar dilihat pada permukaan matriks (SPS) yang disebabkan oleh penyerapan air. Akhirnya, ujian biodegradasi dan luluhawa juga dikaji untuk kedua-dua biopolimer dan biokomposit. Kesan alam sekitar terhadap SPS terplastik dan SPF/SPS biokomposit adalah masing-masing mengalami penurunan kekuatan tegangan sebanyak 78.09% dan 53.67% pada akhir 72 jam tempoh ujian luluhawa. Ujian biodegradasi menunjukkan bahawa SPS terplastik terurai dengan cepat dan kehilangan berat sebanyak 63.58% pada akhir 72 jam berbanding dengan SPF/SPS biokomposit.

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I certify that a Thesis Examination Committee has met on **02 August 2013** to conduct the final examination of **Sahari Japar** on his thesis entitled **"Characterization and development of biopolymer derived from** *arenga pinnata* **and their biocomposites "** 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.

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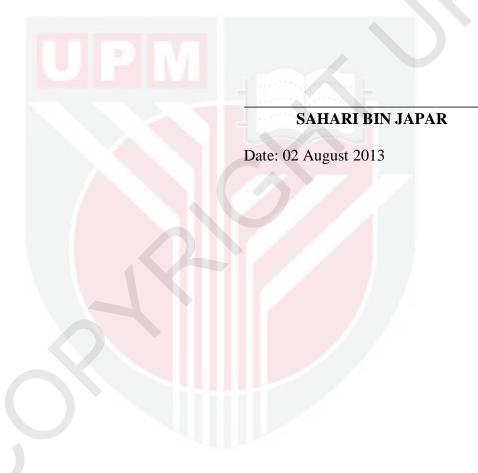
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DECLARATION

I declare that the thesis is my original work except for quotations and citations which have been duly acknowledged. I also declare that it has not been previously and is not concurrently, submitted for any other degree at Universiti Putra Malaysia or at any other institution.



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LIST OF ABBREVIATIONS

ASTM American Society for Testing and Material

AGE Allyl glycidyl ether modified

CO₂ Carbon dioxide

DP Degree of polymerization

DSC Differential scanning calorimetry

DTG Derivative Thermogravimetric Analysis

FTIR Fourier transform infrared spectroscopy

INTROP Tropical Forestry and Forest Products

IRRI International Rice Research Institute

KBr Potassium bromide

MF Medium fibre

LDPE Low density polyethylene

LIPI Lembaga Ilmu Pengetahuan Indonesia

PHBV Poly(3-hydroxybutyrate-co-3-hydroxyvalerate)

PF Phenol formaldehyde

PLA Polylactic acid

RH Relative Humidity

SEM Scanning electron microscope

SF Short fibre

SPB Sugar palm bunch

SPD Sugar palm frond

SPF Sugar palm fibre

SPF/SPS Sugar palm fibre reinforced sugar palm starch

SPS Sugar palm starch

SPT Sugar palm trunk

TAPPI Technical Assn of the Pulp and Paper Industry

TGA Thermogravimetric analysis

TG Thermogravimetric

Tm Melting temperature

Tg Glass transition temperature

TPS Thermoplastic starch

TPRS Thermoplastic rice starch

UP Unsaturated polyester

UV Ultraviolet

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