UNIVERSITI PUTRA MALAYSIA

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Thesis Submitted to the School of Graduate Studies, Universiti Putra Malaysia in Fulfillment of the Requirement for the Degree of Master of Science

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Abstract of the thesis presented to the Senate of Universiti Putra Malaysia in fulfillment of the requirement for the degree of Master of Science

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March 2009

Chairman : Nor Azowa Binti Ibrahim, PhD

Faculty : Science

Natural fiber reinforced composites using thermoplastic such as polypropylene and polyethylene as a matrix produced partially biodegradable composites. In order to produce totally biodegradable composites or green composites, poly(ε-caprolactone) was used as the matrix. However the hydrophilic nature of oil palm empty fruit bunch fiber (natural fiber) affects negatively its adhesion to hydrophobic polymer matrix, thus to improve the compatibility of both components a cross-linker, poly(*N*-vinylpyrrolidone) and electron beam radiation have been proposed. The composites of OPEFB:PCL were prepared by melt blending technique using Haake Internal Mixer. The effect on the amount PVP and doses of electron beam irradiation on mechanical properties of OPEFB:PCL were studied.

The properties of OPEFB:PCL composites were improved by addition of 1% by weight of PVP and irradiated with 10 kGy of electron beam. The FTIR spectra indicate a slight interaction between OPEFB with PCL after adding PVP and irradiation in agreement

with the significant improvement of mechanical properties. The tensile strength of OPEFB:PCL without PVP and treatment is 10.3 MPa whereas after addition of PVP and treatment the tensile strength increased to 16.7 MPa indicating good stress transfer from OPEFB to PCL matrix. Addition of PVP and treatment with electron beam also increase the flexural strength and modulus to 24.32 and 8.69 % respectively. The impact strength is also slightly increased with PVP and irradiation which is about 2.28 %.

From XRD patterns, it can be inferred that the amorphous phase of the composites is slightly increased after adding PVP whereas no significant change was observed after irradiation. Thermal properties of the composites were studied by thermogravimetric analysis (TGA) and differential scanning calorimetry (DSC). From the results, there is also no significant improvement observed for thermal stability of the composites. The surface morphology of the facture surface obtained from tensile test shows no fiber pull out indicating interaction between the OPEFB and PCL after addition of PVP and irradiation.

The environmental degradation behavior on the physical properties of OPEFB:PCL composites has been studied with special reference to the influence of ageing conditions like treatment with water and soil degradation. From water uptake analysis, it can be inferred that the composites become more water resistant after the irradiation. The soil burial test was carried out in 3 months, indicates that irradiation and fiber loading tend to promote degradation of the composites.

Abstrak tesis yang dikemukakan kepada Senat Universiti Putra Malaysia sebagai memenuhi keperluan untuk ijazah Master Sains

BIOKOMPOSIT DARIPADA POLIMER YANG TERBIODEGRADASI DAN TANDAN KOSONG BUAH KELAPA SAWIT (OPEFB) YANG TELAH DIUBAHSUAI

Oleh

SITI NUR AFIFI BINTI AHMAD

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Komposit yang diperkuatkan dengan fiber semulajadi menggunakan termoplastik seperti polipropilena (PP) dan polietelina (PE) sebagai matriks menghasilkan komposit yang separa terbiodegradasi. Bagi menghasilkan komposit yang terbiodegradasi sepenuhnya, polikaprolakton (PCL) telah digunakan sebagai matriks. Walau bagaimanapun sifat hidrofilik fiber tandan kosong buah kelapa sawit (fiber semulajadi) memberi kesan negatif terhadap perlekatan dengan matriks polimer yang hidrofobia, oleh yang demikian, untuk meningkatkan kesesuaian kedua-dua komponen penaut silang, poli(*N*-vinilpirolidon) (PVP) dan radiasi alur elektron digunakan. Komposit OPEFB:PCL disediakan dengan teknik pengadunan lebur menggunakan alat pencampur dalaman ‘Thermo Haake’. Kesan kuantiti PVP dan dos alur elektron yang digunakan terhadap ciri-ciri mekanikal OPEFB:PCL telah dikaji.

Ciri-ciri komposit ditingkatkan dengan penambahan PVP sebanyak 1% dan radiasi alur elektron dengan dos sebanyak 10 kGy. Spektra FTIR menunjukkan sedikit interaksi di antara fiber dan PCL selepas penambahan PVP dan radiasi sejajar dengan peningkatan yang ketara pada ciri-ciri mekanikalnya. Kekuatan tensil OPEFB:PCL tanpa PVP dan rawatan adalah 10.3 MPa manakala selepas penambahan PVP dan rawatan kekuatan tensil meningkat kepada 16.7 MPa yang menunjukkan pemindahan ketegangan yang baik daripada OPEFB kepada matriks PCL. Penambahan PVP dan rawatan dengan alur elektron juga menaikkan kekuatan dan modulus flektural masing-masing daripada 16.8 kepada 22.2 MPa dan 1072 kepada 1174 MPa. Kekuatan kesan hentaman juga meningkat sedikit dengan penambahan PVP dan radiasi iaitu sebanyak 2.28%.

Daripada analisis XRD, ia boleh disimpulkan bahawa fasa tak berbentuk komposit tersebut sedikit bertambah selepas penambahan PVP manakala tiada perubaham yang ketara didapati selepas radiasi. Sifat-sifat terma bagi komposit OPEFB:PCL telah dikaji dengan menggunakan Analisis Thermogravimetri (TGA) dan Analisis kalorimetri pengimbas pembezaan (DSC). Tiada peningkatan kestabilan terma yang jelas pada komposit tersebut yang didapati daripada pemerhatian. Tata bentuk permukaan pada pecahan permukaan ujian tensil menandakan tiada fiber yang tercabut menunjukkan interaksi di antara OPEFB dan PCL selepas penambahan PVP dan radiasi.

Kesan degradasi persekitaran kepada ciri-ciri fizikal dan mekanikal bagi komposit OPEFB:PCL telah dikaji dengan melibatkan ujian sampel ke atas penyerapan air dan degradasi tanah. Merujuk kepada analisis kadar penyerapan air, ia boleh disimpulkan bahawa komposit menjadi lebih tahan kepada serapan air selepas rawatan radiasi. Penanaman sampel dalam tanah dilakukan selama 3 bulan, ia menunjukkan bahawa radiasi dan pemuatan fiber lebih cenderung menggalakkan kepada degradasi komposit.

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I certify that an Examination Committee has met on **24th March 2009** to conduct the final examination of Siti Nur Afifi Ahmad on her Master of Science thesis entitled "**Biocomposites from Biodegradables Polymer and Modified Oil Palm Empty Fruit Bunch Fiber**" in accordance with Universiti Pertanian Malaysia (Higher Degree) Act 1980 and Universiti Pertanian Malaysia (Higher Degree) Regulations 1981. The Committee recommends that the candidate be awarded the relevant degree. Members of the Examination are as follows:

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This thesis submitted to the Senate of Universiti Putra Malaysia and has been accepted as fulfillment of the requirement for the degree of Master of Science. The members of Supervisory Committee are as follows:

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Date: 9 July 2009

DECLARATION

I declare that the thesis is based on my original work except for quotations and citations which have been duly acknowledged. I also declare that it has not been previously or concurrently submitted for any other degree at UPM or other institutions.

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SITI NUR AFIFI AHMAD

Date:

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LIST OF ABBREVIATION/NOTATION

ABS	Acrylonitrile-butadiene-styrene
AS	(3-aminopropyl)-triethoxysilane
ASTM	American Standard for Testing and Materials
BGRP	Bamboo fiber reinforced polypropylene composite
CPE	Chlorinated polyethylene
DMA	Dynamic mechanical analysis
DSC	Differential scanning calorimetry
DTG	Differential thermogravimetry
EFB	Empty fruit bunch
EP-MAH	Maleated ethylene-propylene
EVA	Ethylene/vinyl acetate
FFB	Fresh fruit bunch
FTIR	Fourier transform infrared
GF	Glass fiber
GMA	Glycidyl metacrylate
GP/PP	Glass fiber-polypropylene composite
HDPE	High density polyethylene
LDPE	Low density polyethylene
MAPP	Maleic anhydride maleated polypropylene
MMA	Methyl methacrylate
MPa	Mega Pascal
MPOB	Malaysian Palm Oil Board
MS	Malaysian Standard
OPEFB	Oil palm empty fruit bunch
OPEFB:PP	Oil palm empty fruit bunch-polypropylene composite
PE	Polyethylene
PE/WF	Polyethylene-wood fiber composite
PET	Polyester
PIB	Polyisobutylene

PLA	Poly-lactic acid
PMMA	Polymethyl metacrylate
PORIM	Palm Oil Research Institute of Malaysia
PP	Polypropylene
PP/RNFC	Polypropylene-recycled newspaper cellulose fiber
PP/WF	Polypropylene-wood fiber composite
PPG	Polypropylene glycol
PP-g-GMA	Polypropylene grafted glycidyl metacrylate
PS	Polystyrene
PVC	Poly(vinyl chloride)
PVP	poly(*N*-vinylpyrrolidone)
R&D	Research and development
RNCF	Recycled newspaper cellulose fiber
SBS	Styrene-butadiene-styrene
T_g	Glass transition temperature
TGA	Thermogravimetric analysis
T_m	Crystalline melting point
UV	Ultra-violet
WPC	Wood-plastic composite

CHAPTER 1

INTRODUCTION

1.1 General Background

One major drawback to polymers is the problem of disposal. Since they are somewhat resistive to degradation, polymers tend to accumulate in disposal system, the landfill. Questions about how do polymers and products of biodegradation affect the environment must be taken seriously.

Many solutions have been proposed for soil waste management of plastics, like recycling, incineration, landfill disposal, and degradable plastics. Polymer recycling is an environmentally attractive solution, but the results on a worldwide scale have not been successful because recycling will not yield quality products due to the heterogeneous nature of plastics. Incineration of plastics will release toxic gasses and vapors, which results in a serious health hazard. The use of plastic in landfill operation is least preferred because of space constraint. For these reasons, many investigations have been carried out on the synthesis and manufacture of new polymeric materials which are friendlier to the environment (Abdel-Rehim *et al*., 2004). Consequently, the use of biodegradable polymers such as poly(ε-caprolactone) (PCL) seems to be the best solution to this problem.

Major difficulties in using PCL are poor availability, poor process ability, low toughness, high price and low moisture stability (Chen *et al*., 2005(a); Wang *et al*., 1998). The 60°C melting point of PCL is also too low for many applications. Preparation of blends or conventional composites using inorganic or natural fillers, respectively are among the routes to improve some of the properties of biodegradable polymers.

Lignocellulosic materials, which are among the most environmentally friendly agro wastes were used as reinforcing filler as a substitute for synthetic materials to obtain more biodegradable composites (Hottotuwa *et al*., 2002). Properties and cost of biodegradable polymers can be also modified and improved through the use of lignocellulosic fibers that reduce the cost of the material without modifying their biodegradability (Iannace *et al*., 1999). Furthermore, these materials can be easily obtained from waste products and have a minimal effect on the environment, due to their biodegradable properties. Thus, the emphasis has increasingly been placed on these composites, which may play a major role in resolving some of the pressing environmental issues.

Natural fibers in the form of fiber or/and particulate have been widely used as reinforcing fillers in thermoplastic composites materials. These natural fibers are lighter, and cheaper, decrease the erosion of the manufacturing machinery and provide much higher strength per unit mass than most inorganic fillers. Besides ecological considerations, several technical aspects promote the renewed interest for the fibers as