UNIVERSITI PUTRA MALAYSIA

COIR FIBRE-LOW DENSITY POLYETHYLENE COMPOSITE AS A PACKAGING MATERIAL

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By

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Thesis Submitted to the School of Graduate Studies, Universiti Putra Malaysia, In Fullfilment of the Partial Requirement for the Degree Master of Science

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DEDICATION

To my beloved husband, Mirul;

My loving son, Afiq;

My parent, sisters and brothers.

Thanks for your patience, encouragement and loving support.

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

COIR FIBRE-LOW DENSITY POLYETHYLNE COMPOSITE AS A PACKAGING MATERIAL

By

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July 2002

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Faculty: Engineering

The overall goal of current research was to produce a polymer composite consisting of coir fibre (CF) ie. coconut palm fruit fibre and low density polyethylene (LDPE) that would be suitable for food packaging application. CF and LDPE coupled with and without maleated polyethylene (MAPE) materials have been compounded using a twin screw extruder. The mechanical and physical properties of both composite systems were compared. The effect of fibre and MAPE loading on those properties have also been analyzed. In addition, maleated polyethylene with two different molecular weight (M_w) but having the same acid number have also been incorporated into composites to study the effect of molecular weight on the properties of composites. The incorporation of CF into LDPE matrix has resulted in the positive improvement in the tensile and flexural strength and moduli. Whereas, unnotched impact strength (IS), density, water absorption and thickness swelling decreased as the fibre loading was increased. It was observed that the tensile and flexural strength, unnotched IS and elongation at break of composites have been improved almost doubled that of uncoupled composite. The dimensional stability of composites had also improved after addition of MAPE. Mechanical and physical properties of composites were found to depend strongly on the M_w and sufficient maleic anhydride content of the functionalized polyethylene. Environmental scanning electron microscopy (ESEM) displayed that the adhesion between fibre and matrix had been improved at interfacial region. The ESEM micrographs exhibited evidence that an even fibre distribution might play a dominant role on the properties.

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

KOMPOSIT SERABUT KELAPA-POLIETILENA BERKETUMPATAN RENDAH SEBAGAI BAHAN PEMBUNGKUSAN

Oleh

ROSNITA A. TALIB

Julai 2002

Pengerusi: **Profesor Madya Mohd Nordin Bin Ibrahim, Ph.D.**

Fakulti: **Kejuruteraan**

Tujuan penyelidikan ini dilakukan adalah untuk menghasilkan komposit polimer yang terdiri daripada serabut kelapa (CF) dan polietilena berketumpatan rendah (LDPE) yang sesuai diguna sebagai pembungkus makanan. CF dan LDPE digabungkan bersama bahan atau tanpa bahan maleated polietilena (MAPE) dengan menggunakan penyemperit skru berkembar. Sifat mekanikal dan fizikal kedua-dua sistem komposit ini kemudiannya dibandingkan. Kesan penambahan serabut kelapa dan MAPE terhadap sifat-sifat komposit turut dianalisa. Di dalam kajian ini, MAPE yang mengandungi dua berat molekul yang berbeza tetapi nombor asid yang sama ditambahkan ke dalam komposit untuk menentukan kesan berat molekul terhadap sifat-sifat komposit. Penambahan serabut kelapa ke dalam matriks LDPE menghasilkan peningkatan positif

terhadap kekuatan dan modulus bagi tegangan dan lenturan. Sebaliknya, penambahan serabut kelapa mengakibatkan pengurangan terhadap kekuatan impak tanpa lekuk, ketumpatan, kekuatan, serapan air dan ketebalan pembengkakan. Berdasarkan pemerhatian, didapati kekuatan tegangan dan lenturan, kekuatan impak tanpa lekuk dan pemanjangan pada titik kegagalan telah meningkat hampir dua kali ganda berbanding komposit tanpa bahan penambah. Penambahan MAPE juga meningkatkan kestabilan dimensi komposit. Didapati, sifat-sifat mekanikal dan fizikal amat bergantung terhadap berat molekul dan kandungan maleik anhidrat yang mencukupi bagi polietilena berfungsi. Pengimbas elektron mikroskopi alam sekitar (ESEM) menunjukkan peningkatan terhadap pelekatan antara serabut kalapa dan matrik pada kawasan antara muka. Mikrograf-mikrograf ESEM menunjukkan bukti bahawa penyebaran sekata serabut memainkan peranan besar terhadap sifat-sifat komposit.

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DECLARATION

I hereby 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 instituitions.

ROSNITA A. TALIB

Date:

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LIST OF ABBREVIATIONS

ASTM	American Society for Testing and Materials
A	Area
APE	3-aminopropyltriethoxysilane
APM	3-aminopropyltrimethoxysilane
C 18	Maleated Polyethylene with M_w of 26,000
C16	Maleated Polyethylene with M_w of 15,000
CF	Coir Fibre
EFB	Empty Fruit Bunch
ESCA	Electron Spectroscopy for Chemical Analysis
ESEM	Environmental Scanning Electron Microscpy
F	Force
FS	Flexural Strength
HDPE	High Density Polyethylene
IR	Infrared
IS	Impact Strength
LDPE	Low Density Polyethylene
LLDPE	Linear Low Density Polyethylene
MA	Maleic anhydride
MAH-PP	Polypropylene-maleic anhydride
MAPE	Maleated Polyethylene
MOE	Modulus of Elasticity
MOR	Modulus of Rupture

M_w	Molecular Weight
NMR	Nuclear Magnetic Resonance
OH	Hydroxyl
OP-EFB	Oil Palm- Empty Fruit Bunch
PMMA	p-methoxymethylamphetamine
PMPPIC	Polymethylene-polyphenyl-isocyanate
PP	Polypropylene
PS	Polystyrene
PVC	Polyvinyl chloride
SEM	Scanning Electron Microscpy
SIRIM	Standards and Industrial Research Institute
TEM-EDXA	Transmission Electron Microscopy-Electron Dispersive X-Ray Analysis
T_g	Glass Transition Temperature
TM	Tensile Modulus
TPM	3-trimethoxysilyl-propylmethacrylate
TS	Tensile Strength
TSw	Thickness Swelling
UP	Unsaturated polyester
USM	University Science of Malaysia
VDC	Vinylidene chloride
WA	Water Absorption
WF	Woof Flour

CHAPTER 1

INTRODUCTION

1.1 Overview

Over the past decade, low density polyethylene (LDPE) has become popular in the food packaging industry because it is relatively cheap thermoplastic with superior mechanical properties compared to high density polyethylene. It has an outstanding impact strength, low permeability to water vapour, can be coloured to a variety of shades. In addition, its processability, rescalability and recyclability have made LDPE the packaging of choice for various application in packaging industry, including heavy duty sacks, refuse sacks, carrier bag and for general packaging (Brydson, 1995).

Foods have been packaged in various ways for thousands of years. As civilization developed. people tried to preserve food with salt or spices and by drying. smoking, or cooling. They also learned from experience that packaging could help preserve food by protecting it from harmful environmental factors such as air, moisture and light.

Since the 1930's, the plastic package has assumed a continually increasing role in protective packaging of foods and beverages. The first package, a pouch of vinyl-coated cellophane, proved useful in protecting foods for short periods of time while providing the requisite flexibility, light weight and transparency needed to view contents. Some milestones for plastics used in food packaging are shown in Table 1.1. Clearly, a great many of the most important inventions and advances in plastic science and technology have been directly used in food packaging (Brown, 1992).

1.2 Advantages of Plastics in Food Packages

Plastics have been widely adopted in packages for foods for reasons of their advantages in use compared with other materials. These advantages are reflected in the physical, mechanical and chemical properties of plastics. Foremost among plastic's features providing advantages to food packers are their formability into a practical unlimited range of shapes and wide range of available properties to fit specific applications. Fabricated into finished containers can be accommodated to required shapes and sizes. Properties inherent in synthetic organic polymers can be adjusted, modified or enhanced by formulation or reinforcement and deploying such processing steps as orientation.

Table 1.1: Selected Precursor Developments in the Contemporary Use of Plastics and Other Organic Polymers in Food Packages

PERIOD	DEVELOPMENTS	CONTEMPORARY FOOD USES
1900s	Modified alkyd resin solutions	Can coatings
	Vinyl acetate polymers	Adhesives, films
1910s	Phenol-formaldehyde resins	Closures (e.g., screw caps)
	Cellophane, casting (uncoated)	Lamina in multilayered films
	Viscose sausage casings	Casings for processed meat
1920s	Coated "moistureproof" cellophane	Pouches, dry foods
	Rubber hydrochloride	Films; now in minimal use
	Vinyl chloride polymers. copolymers	Films, blow bottles, can coatings
1930s	Polystyrene	Trays, tubs, lids, foams
	Polyamide (nylon) resins	Barriers in films. sheets. moldings
	Ethylcellulose	Frozen foods hot melt paper coating
	Methylcellulose	Edible films for internal moisture control in foods
	Polyethylene	Films, bottles, thermoforms, heat seals
	Polyvinylidene chloride (PVDC)	Barrier films, multiplayer thermoforms
1940s	Impact polysryrene	Tubs, trays. thermoforms
	Thermoforming of sheet	Trays, tubs; form-fill-seal
	Injection blow molding	Bottles, jars; with threaded closures
	Extrusion blow molding	Large containers, threaded
	Nylon films	Processed meat packs
	Styrene-butadiene latexes	Paper coating. primers
	PVDC lacquer coatings	High barrier cellophane
1950s	Polypropylene	Bottles, structure layers
	Extrusion polyethylene coating on paper board	Milk, juice and frozen food cartons
	Coextrusion	Barrier containers of multiplayer construction
	Polycarbonates	Structures, food bottles. potable water
	Metallized films	Gas and light barrier pouches
	High barrier PVDC coextrusion resins	Multilayer coextrusions of films, thermoformable sheet
	Epoxy-phenolics	Can coatings for many foods
1960s	Polyethylene imine	Dual oven ware: composited with polycarbonates
	Styrene-butadiene resins	Potable water
	Ionomers	Films, coatings, heat seals
	Ethylene copolymers	Films. Coatings, sealant layers
	Polysulphones	Dual oven ware
	Multilayer coextrusion & thermoforming	High barrier food tubs, trays
	Injection molding. reheat and blow process	Pressurized beverage bottles
1970s	Modified polypropylene resins	Multilayer coextrusion adhesives
	Hydrolyzed ethylene-vinyl acetate (EVOH)	Barrier layer in films, thermoforms
	Acrylonitrile resins	Bottles. thermoforms
	Polyethylene terephthalate (PET)	Food bottles. jars, dual ovenware. paperboard tray coatings
	VDC-methylacrylate copolymers	High barrier multiplayer films, containers
	Coinjection blow molding	Retortable barrier tubs
	Ethylene-acrylic acid copolymers	Films adhesives
	Linear low density polyolefins	Moisture barrier films
1980s	Glycol modified PET	Extrusion blow molded foof jars
	Crystallized PET	Dual oven trayware
	Tortuous path barrier blends	Bottles, films
	Amorphous polyamide	Barrier films, containers
	Polyarylamide resin	Barrier films and multilayers; alloys and blends
	Liquid crystal polymers	High temperature, high barrier uses
	Poly (ethylene 2.6 naphthalene dicarboxylate) [PEN]	Monolayer barrier bottles
	Recycle and recovery of plastics	Egg cartons from PET regrind
	Ultra low density polyethylenes	Thin, tough films, e g . for bag-in-box

Source: Brown. 1992

1.3 The Need for New Plastic Composites from Renewable Sources

The use of polymeric materials in food packaging is increasing not only in respect to proportion of packages totally or partially made with these materials but also in the number of polymer types now used in packaging design. Some of the properties that make polymers attractive for package manufacture are lightness, flexibility, chemical resistance, versatility and a wide range of formulation that allow the development of new designs (polymer alone or combined with other materials) to meet the requirement of the particular foodstuff (Jenkins and Harrington, 1991).

In recent years, significant efforts have been made to manufacture thermoplastic composites using agrowaste and agroforest materials such as sawdust, wood fibres, sisal and baggase. The rationale behind these efforts are that the use of these materials as filler in polymeric matrix offer a high specific stiffness and strength, low density and low cost per volume basis. Medium and high density natural fibre composites can be used for small containers, for example, in the tea industry and for large sea-going containers for commodity goods. Unfortunately, the incompatibility between lignocellulosic materials and many polymeric matrices affect the degree of dispersion of the fibres in the matrix and the overall homogeneity of the composite structure (Marcovich *et al*., 1998). Due to this problem, studies on the effectiveness of coupling agents are required to improve the interfacial adhesion between fibre-matrix phases, and consequently, produce composites with better properties.

1.4 Objectives

The objective of this work is to develop coir fibre-LDPE composite coupled with maleated anhydride-PE for food packaging application by using twin screw extrusion technique. This study was emphasized on the mechanical and physical properties of coupled and uncoupled composite system. Thus, to achieve this objective, the work was divided into four scopes. First, to determine the effect of fibre loading on uncoupled composite. Three percentages of fibre loading (by weight) were employed. Second, the effect of MAPE loading on composite was investigated. The comparison between coupled and uncoupled composite was done to study the change in trend when three MAPE loading were applied into the composite system. Third, in order to study the influence of different M_w on the properties of coupled composite, two different M_w of MAPE were employed in this work. In the final scope, the fractured tensile surfaces were observed by using Environmental scanning electron microscope (ESEM) to gain evidences of improvement through the mode of failure, dispersion of fibre in the matrix and interfacial adhesion between fibre/matrix.