

PROPERTIES AND CHARACTERIZATION OF DEGRADABLE LOW
DENSITY POLYETHYLENE/RICE BRAN COMPOSITE FILMS

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A thesis submitted in fulfilment of the
requirements for the award of the degree of
Master of Engineering (Polymer)

Faculty of Chemical Engineering
Universiti Teknologi Malaysia

APRIL 2015

To my beloved parents and friends

ACKNOWLEDGEMENT

First and foremost, I would like to express my sincere gratitude to my supervisor, Dr. Zurina binti Mohamad for her continuous support during my research, for her patience, motivation, enthusiasm, and immense knowledge. Her guidance has helped me in all time of research and during writing this thesis. I would also like to take this opportunity to thank my co-supervisor, Assoc. Prof Dr. Abdul Razak bin Rahmat for his motivation, helpful comments and ideas.

I would also like to express my sincere thanks to all lecturers, technicians and staff of Faculty of Chemical Engineering, UTM and especially from the Department of Polymer Engineering, UTM whom have directly and indirectly assisted me in the experimental works and the preparation of the thesis.

I thank my fellow laboratory friends and technicians Al-Hafiz Ibrahim, Abang Shidi, Zaidi, Nuha Awang, Rynda, Nor Zalina, En. Izzat and En. Suhee Tan for being such the continuous supports and motivations.

Last but not least, I would like to thank my beloved family especially my father Saadan bin Ahmad and my sister and brothers for their inspirations and endless love during this research.

ABSTRACT

This research work aimed to study the mechanical, morphological, thermal, physical and biodegradability properties of low density polyethylene (LDPE)/rice bran (RB) films. LDPE/RB films were prepared using 0-5 wt% RB, glycerol as plasticizer, polyethylene-grafted-maleic anhydride (PEMAH) and Ultra-Plast™ TP10 (TP10) as compatibilizer by twin screw extrusion followed by blown film machine. It was found that the presence of 5 wt% RB contents had an adverse effect on the tensile strength of LDPE/RB films while the incorporation of PEMAH, TP10 and glycerol has increased the tensile strength of LDPE/RB films. The scanning electron microscope (SEM) micrographs showed that RB particles were clearly seen on the surface of all LDPE/RB films while addition of glycerol, PEMAH and TP10 in the films increased the distribution and dispersion phase of RB in LDPE matrix. Differential scanning calorimetry (DSC) results showed that degree of crystallinity (X_c) and melting enthalpy (ΔH_m) of the films have increased with the presence of RB content while melting temperature (T_m) and crystallization temperature (T_c) had showed no significant changes for all formulations. The reduction of X_c and ΔH_m was observed upon addition of compatibilizers and plasticizer. Thermogravimetry analysis (TGA) analysis showed that the thermal degradation property slightly increased with the incorporation of RB but the reduction was observed with incorporation of PEMAH, TP10 and glycerol in LDPE/RB films. The incorporation of RB in LDPE has influenced the rate of water absorption and it was significantly increased with the addition of glycerol in LDPE/RB films. The addition of RB in LDPE/RB films has increased their biodegradabilities due to the attack of microorganism in soil while the incorporation of TP10, PEMAH and glycerol in LDPE/RB films also gave significant effects to the rate of biodegradation. The overall results showed that the incorporation of PEMAH and glycerol in LDPE/RB films performed better properties especially their mechanical properties.

ABSTRAK

Penyelidikan ini bertujuan untuk mengkaji sifat-sifat mekanikal, morfologi, haba, fizikal dan bioterurai filem polietilena berketumpatan rendah (LDPE)/dedak padi (RB). Filem LDPE/RB telah disediakan dengan menggunakan 0-5 wt% RB, gliserol sebagai pemplastik, polietilena graf maleik anhidrida (PEMAH) dan Ultra-Plast™ TP10 (TP10) sebagai pengserasi dan dihasilkan melalui mesin penyemperit skru berkembar kemudian diikuti mesin peniupan filem. Ini menunjukkan bahawa kehadiran 5 wt% RB telah mengurangkan kekuatan tegangan filem LDPE/RB sementara campuran PEMAH, TP10 dan gliserol telah meningkatkan kekuatan tegangan filem LDPE/RB. Mikrograf mikroskop elektron pengimbas (SEM) menunjukkan bahawa zarah RB dapat dilihat dengan jelas pada permukaan semua filem LDPE/RB sementara itu penambahan gliserol, PEMAH dan TP10 ke dalam filem telah meningkatkan fasa pengagihan dan penyebaran RB di dalam matriks LDPE. Analisis pengimbas pembezaan kalorimeter (DSC) menunjukkan kadar penghabluran (X_c) dan entalpi lebur (ΔH_m) filem telah meningkat dengan kehadiran kandungan RB manakala suhu lebur (T_m) dan suhu penghabluran (T_c) tidak menunjukkan sebarang perubahan ketara untuk semua formulasi. Penurunan X_c and ΔH_m dapat dilihat melalui penambahan pengserasi dan pemplastik. Analisis thermogravimetri (TGA) menunjukkan degradasi haba mengalami sedikit peningkatan apabila RB dicampurkan tetapi berlakunya penurunan apabila PEMAH, TP10 dan gliserol dicampur ke dalam filem LDPE/RB. Campuran RB dalam LDPE mempengaruhi kadar penyerapan air dan ia meningkat lebih ketara dengan penambahan gliserol di dalam filem LDPE/RB. Penambahan RB ke dalam filem LDPE/RB telah meningkat biodegradasi disebabkan serangan mikroorganisma tanah manakala campuran TP10, PEMAH dan gliserol dalam filem LDPE/RB telah membawa kesan yang besar ke atas kadar biodegradasi. Keputusan menyeluruh menunjukkan campuran PEMAH dan gliserol ke dalam filem LDPE/RB boleh menghasilkan sifat-sifat yang terbaik terutamanya sifat mekanikal.

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LIST OF ABBREVIATIONS

ABS	-	Acrylonitrile-butadiene-styrene
DCP	-	Dicumyl peroxide
DSC	-	Differential scanning calorimeter
HDPE	-	High density polyethylene
HV	-	Hydroxyvalerate
LDPE	-	Low density polyethylene
LLDPE	-	Linear low density polyethylene
MAH	-	Maleic anhydride
mcl	-	Medium chain length
MDPE	-	Medium density polyethylene
MFI	-	Melt flow index
MLDPE	-	Maleated low density polyethylene
PA	-	Polyamide
PBS	-	Poly(butylenes succinate)
PCL	-	Poly(ϵ -caprolactone)
PE	-	Polyethylene
PEG	-	poly(ethylene glycol)
PET	-	Polyethylene terephthalate
PEMAH	-	Polyethylene grafted maleic anhydride
PHA	-	Polyhydroxyalkanoate
PHB	-	Poly-3-hydroxybutyrate
phr	-	Part per hundred resin
PLA	-	Poly(lactic acid) or Poly(lactide)
PP	-	Polypropylene
PS	-	Polystyrene
PVA	-	Poly(vinyl acetate)

PVC	-	Polyvinyl chloride
RB	-	Rice bran
scl	-	Short chain length
SEM	-	Scanning electron microscope
TGA	-	Thermogravimetry analysis
TP10	-	Ultra-plast TM TP10
TPS	-	Thermoplastic starch
WVPR	-	Water vapor transmission rate

LIST OF SYMBOLS

cm	-	Centimeter
g	-	Gram
kg	-	Kilogram
ml	-	Milliliter
mm	-	Millimeter
MPa	-	Mega Pascal
CH ₄	-	Methane
CO ₂	-	Carbon dioxide
N ₂	-	Nitrogen
T _c	-	Crystallization temperature
T _g	-	Glass transition temperature
T _m	-	Melting temperature
Wt %	-	Weight percent
X _c	-	Degree of crystallinity
°C	-	Degrees celsius
°F	-	Fahrenheit
ΔH _m	-	Melting enthalphy
μm	-	Micrometer
%	-	Percent

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CHAPTER 1

INTRODUCTION

1.1 Background of Study

Since 1950, almost 300 million tons of plastics have been used around the world for various applications in industry, as reported by Plastic Europe in 2008. Plastics are widely used due to their properties such as light weight, low cost, good mechanical properties and easy processibility (Khodaverdi *et al.*, 2014). However, it is a problem to manage the plastic waste because it takes a long time to degrade or cannot be degraded properly. Some plastics can be degraded into substances but they generate or release hazardous and polluting substances. Therefore, they adversely affect the environment, economical and ecological system especially to human being. Usually, most plastic wastes are dumped and left to degrade naturally (Nagarajan *et al.*, 2012). As a result, the availability of landfill area has decreased dramatically while the cost to provide for landfilling waste has increased drastically. Therefore, recycling and incineration methods have been practiced to minimize these problems. However, only a small percentage of plastic waste is recyclable and most of them end up in the municipal landfill. On top of that, emission of corrosive gas, high capital cost, toxic gases and operation at high temperature make incineration less attractive (Shah *et al.*, 1995).

One of the solutions to tackle the problem of plastic waste management is the production and use of environmentally friendly degradable polymers especially in the packaging applications (Torres *et al.*, 2008). Normally, there are many renewable

sources used to produce biodegradable polymer product such as starch, chitosan, wool and silk. The advantages of using biodegradable product are that they are relatively inexpensive and environmentally friendly. There have been demands to use biodegradable polymer that are comparable to substitute the growing use of non-biodegradable polymer (Tudorachi *et al.*, 2000). Biodegradable polymers are very useful for applications in medical, agriculture, drug release and packaging.

Polyethylene (PE) is a thermoplastic commodity produced by the chemical industry and always used for various application fields. PE is the simplest polymer with just two carbons and four hydrogens in the basic polymer repeating unit. Among the polyolefins, low density polyethylene (LDPE) is more susceptible to the attack of microorganisms in determined conditions (Ohtake *et al.*, 1998). Therefore, LDPE is used to be incorporated with various fibers to produce degradable polymer. According to Girija and Sailaja (2006), LDPE has the largest tonnage in the world, and disposal of this plastic waste has become a major environmental hazard. Hence, attempts to develop degradable blends have been the focus of researches (Anuradha and Kumar, 1999; Yu *et al.*, 2006; Kasirajan and Ngouajio, 2012). Therefore, incorporation of rice bran in polyethylene was expected to attract microorganisms to attack the rice bran content in the polyethylene blends. Furthermore, rice bran is a good choice because of its mass production and low cost in the market, thus will reduce the cost of production of LDPE/rice bran as a degradable polymer film.

Today, degradable plastic is very popular which will decompose in nature either in aerobic or anaerobic condition. Degradable polymer is a product that can be degraded when microorganisms such as fungi, algae and bacteria attack the polymer chains. Usually, pure polymer could become more degradable with the incorporation of additive such as rice bran and starch. The major role of starch has been found to provide higher oxygen permeability as it is consumed by microorganisms (Rutkowska *et al.*, 2002). The matrix is hollowed out and the surface/volume ratio increases. The incorporation of natural filler such as starch had decreased their mechanical properties. Therefore, the compatibilizers and plasticizer were used to improve the properties. Types of compatibilizers that are used to improve the compatibility between two immiscible phases and enhance the mechanical properties

of the blends (Bikiaris and Panayiotou, 1998) such as polyethylene – grafted-maleic anhydride (PEMAH) and Ultra Plast™ TP10. While the plasticizer such as glycerol was needed to enhance the flexibility, softness, resilient and easiness to process. The incorporation of glycerol in polymer/natural filler blends also enhances the distribution and dispersion phase of natural filler in the polymer matrix.

1.2 Problem Statement

Today, the challenge of producing degradable polymer becomes the focus of research interest in order to overcome the problem of plastic waste management. Many researches have been carried out to produce degradable plastic films such as blending of LDPE and materials from natural resources such as starch (Garg and Jana, 2006; Girija and Sailaja, 2006), flour (Morreale *et al.*, 2008) and chitosan (Bourtoom and Chinnan, 2008). With the addition of natural filler or fibre the biodegradability has been improved and the product has become more environmentally friendly. In this study, rice bran from local producer was used to produce LDPE/rice bran film. According to George *et al.* (2006), biodegradability can be increased with increasing rice bran content in polyethylene films. In this study, the particle size of the rice bran was set in the range of 100 – 500 µm. The tensile properties, thermal, biodegradability, physical and morphology were evaluated.

This study aims to determine the properties and characteristics of degradable film based on LDPE/ rice bran films. In this study several questions which need to be answered are as follows:

- i) What was the maximum ratio of rice bran (RB) loading that could be processed to produce degradable LDPE films?
- ii) What was the effect of rice bran loading on the properties of degradable LDPE/RB films?

- iii) Did addition of rice bran improve the biodegradability of degradable LDPE films?
- iv) What was the effect of different types compatibilizer and plasticizer such as polyethylene maleic anhydride (PEMAH), Ultra PlastTM TP10 and glycerol on the tensile properties, characterization and biodegradability for LDPE/RB films?

1.3 Objectives of the Study

This research was aimed to produce degradable film based on polyethylene blend with rice bran via extrusion and blow film machine.

Specific objectives of this work are:

- i) To determine the maximum processable ratio of filler loading (rice bran) on degradable LDPE/RB film.
- ii) To investigate the effect of different rice bran contents on the properties such as mechanical properties, morphology, thermal properties, water absorption and biodegradability of degradable LDPE/RB films.
- iii) To study the effect of compatibilizer and plasticizer such as PEMAH, TP10 and glycerol on the properties and the characterization of degradable LDPE/RB films.

1.4 Scope of the Study

The scopes of this study are:

- i) Preparation of sample by using twin screw extruder and blown film machine.
- ii) Tensile test to evaluate the mechanical properties of the films.
- iii) Scanning electron microscope (SEM) to study the morphology of these films.

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