

**EFFECT OF FILLER LOADING AND CHEMICAL
MODIFICATION ON PROPERTIES OF LOW
DENSITY POLYETHYLENE/CORN STALK
BIOCOMPOSITES**

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UNIVERSITI MALAYSIA PERLIS

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DENSITY POLYETHYLENE/CORN STALK
BIOCOMPOSITES**

by

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LIST OF ABBREVIATIONS

ASTM	American Society for Testing and Materials
ATR	Attenuated total reflectance
CAPE	Carboxylated polyethylene
CS	Corn Stalk
COCA	Coconut Oil Coupling Agent
DMA	Dynamic mechanical analysis
DPW	Date palm wood
DSC	Differential scanning calorimetric
EDD	Ethylene diamine dilaurate
FTIR	Fourier transform infrared spectroscopy
HDPE	High density polyethylene
HPLC	High performance liquid chromatography
LDPE	Low density polyethylene
LLDPE	Linear low density polyethylene
MA	Maleic Anhydride
MAPE	Maleated polyethylene
MAPP	Maleated polypropylene
NFPC	Natural fiber polymer composite
OHF	Olive Husk Flour
PALF	Pineapple leaf fiber
PE	Polyethylene

PET	Polyethylene terephthalate
PE-g-MA	Polyethylene grafted maleic anhydride
PKS	Palm kernel shell
POFA	Palm oil fatty acid
PP	Polypropylene
PP-g-MA	Polypropylene grafted maleic anhydride
RHDPE	Recycled high density polyethylene
SEM	Scanning electron microscopy
TDM	Titanium derived mixture
TGA	Thermogravimetric analysis
TPRS	Thermoplastic rice starch
TS	Tensile strength
WOWP	Waste office white paper
WP	Waste paper
XPS	X-ray photoelectron spectroscopy

LIST OF SYMBOLS

M_t	Percentage of water absorption
T_c	Crystallization temperature
T_g	Glass transition temperature
T_m	Melting point temperature
W_d	Original dry weight
W_n	Weight after immersed
X_{biocom}	Crystallinity of biocomposites
X_c	Degree crystallinity of eco-composites

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Kesan Pembebanan Pengisi Dan Modifikasi Kimia Ke-Atas Sifat-Sifat Biokomposit Polietilena Ketumpatan Rendah/Batang Jagung

Abstrak

Di dalam kajian ini, penggunaan batang jagung (BT) sebagai pengisi di dalam polietilena ketumpatan rendah (PEKR) telah dikaji. Kesan pembebanan pengisi BT dan modifikasi kimia ke atas sifat-sifat kekuatan tensil, morfologi, penyerapan air dan sifat-sifat terma biokomposit PEKR/BT telah dikaji. Tiga jenis-jenis modifikasi kimia telah digunakan, seperti maleik anhidrida polietilena (MAPE), agen pengganding kelapa (APK) dan eko-rosotan PD-04. Biokomposit disediakan menggunakan *Brabender Plasticorder EC PLUS* pada suhu 160⁰C dan kelajuan rotor 50 rpm. Keputusan menunjukkan bahawa dengan semakin meningkatnya pembebanan pengisi BT didapati kekuatan tensil dan pemanjangan pada takat putus biokomposit PEKR/BT berkurang, di mana modulus Young dan penyerapan air didapati meningkat. Morfologi patahan permukaan tensil biokomposit menunjukkan pelekatan dan interaksi antara muka yang lemah diantara pengisi hidropilik BT dan hidropobik matrik PEKR. Biokomposit PEKR/BT pada kandungan BT 20 bsp menunjukkan penghabluran yang paling tinggi diikuti PEKR tulen dan biokomposit dengan pembebanan BT 40 bsp. Jumlah pengurangan berat biokomposit berkurang dengan meningkatnya kandungan pengisi. Ini menunjukkan pada suhu yang lebih tinggi biokomposit mempunyai ketahanan terma yang lebih baik. Modifikasi-modifikasi kimia dengan MAPE, APK dan eko-rosotan telah meningkatkan sifat-sifat tensil dan terma biokomposit. Kehadiran MAPE telah meningkatkan kekuatan tensil dan modulus Young biokomposit dengan pengserasi, tetapi pemanjangan pada takat putus didapati berkurang. Biokomposit terawat dengan APK atau eko-rosotan mempunyai kekuatan tensil dan pemanjangan takat putus yang lebih tinggi, manakala modulus Young yang lebih rendah dibandingkan biokomposit tanpa rawatan. Biokomposit terawat dengan MAPE, APK dan eko-rosotan menunjukkan ketahanan terhadap penyerapan air yang lebih baik daripada biokomposit tanpa rawatan. Didapati penghabluran dan kestabilan terma biokomposit terawat adalah lebih tinggi berbanding biokomposit tanpa rawatan. Kajian SEM biokomposit yang dirawat dengan MAPE, APK dan eko-rosotan telah meningkatkan interaksi antara muka BT dan matrik PEKR. Kehadiran MAPE, APK dan eko-rosotan di dalam biokomposit telah dibuktikan dengan kumpulan berfungsi yang baru pada spektra FTIR.

Effect Of Filler Loading And Chemical Modification On Properties Of Low Density Polyethylene/Corn Stalk Biocomposites

Abstract

In this study, the utilization of corn stalk (CS) as a filler in low density polyethylene (LDPE) was investigated. The effect of CS loading and chemical modification on tensile properties, morphology, water absorption and thermal properties of LDPE/CS biocomposites were studied. The three types of chemical modification were used, such as maleic anhydride polyethylene (MAPE), coconut coupling agent (COCA), and eco-degradant PD-04. The biocomposites were prepared using Brabender Plasticorder EC PLUS at temperature 160 °C and rotor speed 50 rpm. The results showed that the increased of CS loading caused decreased in the tensile strength and elongation at break of LDPE/CS biocomposites, whereas the Young's modulus and water absorption increased. The morphology of tensile fracture surface of biocomposites showed the poor adhesion and interfacial interaction between hydrophilic CS and hydrophobic matrix. The LDPE/CS biocomposites at 20 php CS loading indicated highest crystallinity followed pure LDPE and biocomposites at 40 php CS loading. The total weight loss biocomposites decreased with increases CS loading. This indicates at higher temperature the biocomposites have better thermal stability. The chemical modifications with MAPE, COCA or eco-degradant had enhanced the tensile and thermal properties of biocomposites. The presence of MAPE has increased the tensile strength and Young's modulus of compatibilized biocomposites, but elongation at break decreased. The treated biocomposites with COCA or eco-degradant have higher tensile strength and elongation, while Young's modulus lower compared to untreated biocomposites. The treated biocomposites with MAPE, COCA and eco-degradant exhibit better water resistance than untreated biocomposites. It was found the crystallinity and thermal stability of treated biocomposites higher than untreated biocomposites. The SEM study of treated biocomposites with MAPE, COCA and eco-degradant showed an enhanced interfacial interaction between CS and LDPE matrix. The presence of MAPE, COCA and eco-degradant in biocomposites were evident by the new functional group from FTIR spectra.

CHAPTER 1

INTRODUCTION

1.1 Research background

Biocomposites from plant derived fiber (natural/biofiber) and crop derived plastics (bioplastic) are novel materials of the twenty-first century and would be a great importance to the materials world, not only as a solution to growing environmental threat but also as a solution to the uncertainty of petroleum supply (Mohanty et al., 2002). The development of natural fibers such as kenaf, flax, jute, hemp and sisal has attracted a lot of researchers because of their advantage in that they can be used in a variety of applications and their effectiveness is similar to traditional fillers such as carbon, aramid and glass (Takasu et al., 2002; Thakur et al., 2011). The ease of processing, cost reduction and productivity are the most significant advantages that the polymers offers other materials (Wang et al., 2003; Murugan et al., 2004; Kuboki et al., 2007). The advantages of natural fiber/filler are low cost, low density, low health hazards, biodegradability, better wear resistance and high degree of flexibility, renewability, and high specific strength (Panthapulakkal et al., 2006; Jia et al., 2007). These biocomposites are being extensively used for the production of cost effectively ecofriendly biocomposites.

It is observed that natural fibers/fillers have properties similar to traditional synthetic fiber reinforced biocomposites (Nagaito & Yano, 2005; Bhatnagar & Sain, 2005). A number of significant industries such as the automotive, construction or

packaging industries have shown massive interest in the progress of new biocomposites materials. In automotive industry, Mercedes Benz has forge ahead against the rest in the industry by using jute reinforced plastic for the interior door panels of its E-class vehicles because of lower cost and lower density. All these properties have made natural fibers/fillers very attractive for various industries currently engaged in searching for new and alternate products to synthetic fiber reinforced biocomposites (Singha & Thakur, 2008).

Biocomposites role in variety of applications is very domineering for a long period of time now due to their specific strength and modulus (Cao et al., 2006). Biocomposites will become commercial application of the future that would unravel the potential of these underutilized renewable materials and offer a non food based market for agricultural industry (Alemdar & Sain, 2008).

The polyolefins such as polypropylene (PP) and polyethylene (PE) have been widely used as synthetic polymers in the commercial plastic industry but their non biodegradability and consequently waste disposal problem in nature environment have caused various forms of environmental pollution (Kim et al., 2006; Kim & Kim, 2008). Plastic matrix which comes from a group of polyethylene thermoplastic has been used broadly in daily life (Pollanen et al., 2013). Low density polyethylene resins are once again known as valuable product family. Its combination of superior clarity with a stiffness and density is much preferred by converters for down gauging. Low density polyethylene (LDPE) is commonly used for manufacturing various containers, dispensing bottles, wash bottles, tubing, plastic bags for computer components and packaging applications (Yang et al., 2007).

Corn stalk is one agriculture crops that is widely cultivated around the world and greater weight of corn produced each year. After harvesting of corn, the residue like leaves, cob, stalk and husks were left as part of corn stover in the field (Yeng et al., 2013). The utilization of corn stalk has potential to be incorporated into value product for plastic industry as natural filler.

Some of the researchers has been reported the utilization of lignocellulosic material or natural filler and thermoplastics group as matrices such as LDPE with doum (Arrakhiz et al., 2013), LDPE with palm kernel shell (Salmah et al., 2011b), LDPE with kenaf (Behjat et al., 2010), HDPE with flax fiber (Li et al., 2009), PE with curaua (Araujo et al., 2008), HDPE with bamboo flour (Liu et al., 2008), LLDPE with wood flour (Kuan et al., 2006), PE with sisal (Torress et al., 2005), and PE with wood flour (Farid et al., 2002).

In general, natural fibers provide many advantages for biocomposites but they pose a problem as the usually polar fibers have inherently low compatibility with non polar polymer matrices, especially hydrocarbon matrices such as polypropylene (PP) and polyethylene (PE). The incompatibility may cause problems in composite processing and material properties. To overcome this incompatibility problem, various physical and chemical methods have been employed to modify the natural fibers (Bledzki & Gassan, 1999). There have been several chemical modification used in earlier research such as by grafting polymers onto the fibers (Xie et al., 2010; Beg & Pickering, 2008), treatment with silane (Girones et al., 2007; Xie et al., 2010), bleaching (Aisaeed et al., 2013), and alkali treatment (Alawar et al., 2009; Roy et al., 2012). One study uses a compatibilizer and coupling agent. The addition of MAPP and MAPE as compatibilizing agents has been used in polyolefins such as polypropylene and

polyethylene with date palm wood (Seyed et al., 2014), chitosan (Azieyantie & Salmah, 2014), corn shell (Li et al., 2013), sisal (Kaewkuk et al., 2013) biocomposites. A compatibilizing agent or coupling agent was used in this study in order to reduce the hydrophilicity of the fillers. The development of fiber dispersion and surface adhesion to polymer to improve stress transmission has become the subject of extensive research.

1.2 Problem Statement

Currently, most of the materials which are produced by fossil fuel and greatly used in packing industries are normally non-degradable. They claimed that as packaging materials for foodstuff, or short term packaging materials, they can create serious issues with environment. By reducing packaging waste, it has encouraged the discovery of new bio-based packaging materials for example biocomposites and biodegradable films from resources that have been renewed. It is hoped that by using all these materials, it can somehow provide a solution the waste problem. The research studies based on natural fiber/filler in thermoplastics biocomposites have been reported by previous study. The utilizations of corn stalk in low density polyethylene (LDPE) has not been reported. Corn stalk is an abundant agriculture waste and potential as filler in thermoplastic biocomposites. The hydrophilic nature of natural fibers affects adhesion to a hydrophobic matrix and as result, there may cause a loss of strength. The mechanical properties of natural filler filled polymer composites depend not only on the properties of constituents but also the degree of interfacial adhesion between the filler and polymer matrix. Poor interfacial bonding leads to biocomposites exhibiting rather poor durability and toughness. In this study, the various type of chemical modifications