CHARACTERISATION AND PROPERTIES OF BIOCOMPOSITES FABRICATED FROM CYPERUS ODORATUS AND LINEAR LOW DENSITY POLYETHYLENE

NIK AHMAD FARIS BIN NIK ABDULLAH

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By

NIK AHMAD FARIS BIN NIK ABDULLAH
1340411112

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UNIVERSITI MALAYSIA PERLIS

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LIST OF ABBREVIATIONS

ASTM	American Society for Testing and Materials
CS	Cobalt Stearate
CY	Cyperus Odoratus
DSC	Differerential Scanning Calorimetry
EB	Electron Beam
Eb	Elongation at Break
FTIR	Fourier Transform Infrared Spectrometer
LLDPE	Linear Low Density Polyethylene
SEM	Scanning Electron Microscope
TGA	Thermogravimetric Analysis
TMPTA	Trimethylolpropane Triacrylate
TPGDA	Tripropylene Gylcol Diacrylate

LIST OF SYMBOLS

ΔH_m	Melting enthalpy
ΔH_f^o	Heat of fusion
ΔH_f^*	Heat of fusion for semicrystalline
phr	Part per hundred resin
$T_{-5\%}$	Temperature at 5% degradation
$T_{-30\%}$	Temperature at 30% degradation
Tc	Crystalline temperature
Tm	Melting temperature
W_i	Weight before degradation
W_f	Weight after degradation
wt%	Percentage in weight
Xc	Degree crystallinity

Pencirian dan Sifat Biokomposit yang dihasilkan daripada *Cyperus Odoratus* dan Polietilena Ketumpatan Rendah Linear

ABSTRAK

Kajian ini dilakukan terhadap potensi *Cyperus Odoratus* (CY) sebagai pengisi. Serbuk CY dicampurkan dengan polietilena ketumpatan rendah linear (LLDPE), sebelum dimasukkan ke dalam penyemperit skru kembar dan seterusnya ke dalam mesin pengacuan suntikan untuk menghasilkan spesimen dumbel biokomposit LLDPY/CY. Biokomposit LLDPE/CY dengan nisbah komposisi 5% hingga 20% berat dikaji. Hasil ujikaji menunjukkan peningkatan pemuatan CY dan saiznya menyebabkan nilai modulus keanjalan bertambah, tetapi kekuatan tegangan dan pemanjangan pada takat putus (Eb) berkurang. Selain itu, pencirian morfologi permukaan patah tegangan bagi biokomposit ini yang ditentukan melalui SEM menunjukkan rekatan antara muka yang lemah antara pengisi CY dan matriks LLDPE termoplastik. Rawatan 5% NaOH terhadap CY meningkatkan kekuatan tegangan dan modulus Young bagi adunan LLDPE/CY dengan ketara, walaupun ini menyebabkan penurunan dalam Eb. Daripada segi penghabluran dan kestabilan haba, biokomposit yang dirawat adalah lebih baik berbanding biokomposit yang tidak dirawat. Sementara itu, penyinaran alur elektron (EBI) dikenakan pada komposit untuk tujuan rangkaian silang, menggunakan pemecut alur elektron 1.5 MeV, dalam julat dos 0–150 kGy. Berdasarkan keputusan kekuatan tegangan, penyinaran pada 100 kGy menghasilkan penyerapan optimum bagi biokomposit LLDPE/CY. Trimetilolpropana triakrilat (TMPTA) dan tripropilena glikol diakrilat (TPGDA) digunakan sebagai pemula dalam kajian ini dan hasil ujikaji menunjukkan komposit yang ditambah dengan TMPTA menunjukkan sifat tegangan, kestabilan terma, kandungan gel, dan penghabluran lebih tinggi, berbanding komposit yang ditambah dengan TPGDA. Hasil kajian juga menunjukkan penambahan TMPTA dan EBI pada 100 kGy meningkatkan semua sifat fizikal, dengan menyediakan bahan yang sesuai berasaskan polimer semula jadi biokomposit. Kehadiran TMPTA meningkatkan rangkaian silang LLDPE/CY semasa penyinaran, yang kemudiannya meningkatkan kestabilan haba komposit tersebut. Timbusan tanah semula jadi dan luluhawa dikenakan ke atas biokomposit LLDPE/CY selama 1 tahun dan sampel diambil selepas 3, 6, dan 12 bulan untuk ujian kebolehuraian, melalui ujian tegangan, kajian morfologi, dan ukuran kesusutan berat sampel. Dalam tempoh 1 tahun ujian penimbusan tanah dan luluhawa, kekuatan tegangan dan Eb bagi komposit menurun, manakala modulus Young meningkat. Kemerosotan dalam sifat-sifat berat komposit dikaji, dan didapati kehadiran pengisi CY dalam komposit mempercepat degradasi biokomposit LLDPE/CY dengan ketara. Kesan kobalt stearat (CS) terhadap penimbusan tanah dan luluhawa ke atas sifat-sifat biokomposit LLDPE/CY turut diselidiki, dan hasil ujikaji menunjukkan kekuatan tegangan dan Eb bagi komposit menurun, manakala nilai modulus Young meningkat dengan bertambahnya kandungan CY dan CS. Pencirian morfologi SEM membuktikan penambahan CS pada komposit mempercepat degradasi.

Characterisation and Properties of Biocomposites Fabricated from *Cyperus Odoratus* and Linear Low Density Polyethylene

ABSTRACT

The potential of *Cyperus Odoratus* (CY) as a filler was studied. In powder form, CY was mixed with linear low density polyethylene (LLDPE), prior to being fed into a twin screw extruder and subsequently into an injection moulding machine to produce LLDPY/CY biocomposite dumbbell specimens. LLDPE/CY biocomposites with composition ratios of 5% to 20 wt% were studied. The results obtained showed that the increased of CY loading and size resulted in an increment of the Young's modulus, but slightly reduction in tensile strength and elongation at break (Eb). In addition, as determined through the SEM, the morphology characterisation of tensile fracture surface of these composites showed poor interfacial adhesion between the CY filler and thermoplastic LLDPE matrix. A 5% NaOH treatment on CY improved the tensile strength and Young's modulus of the LLDPE/CY biocomposites significantly, although it caused a decrement in Eb. The NaOH treatment enhanced both the tensile and thermal properties of the composites. In terms of crystallinity and thermal stability, the treated composites were superior compared with those of the untreated composites. Meanwhile, electron beam irradiation (EBI) was applied on the composites for crosslinking purposes, using a 1.5 MeV electron beam accelerator within the dosage range of 0–150 kGy. Based on the results of the tensile strength, the radiation at 100 kGy caused the optimum absorption by the LLDPE/CY biocomposites. Trimethylolpropane triacrylate (TMPTA) and tripropylene glycol diacrylate (TPGDA) were used as the crosslink promoters and the results indicated that the composites added with TMPTA showed higher tensile properties, thermal stability, gel content, and crystallinity, compared with those of the composites added with TPGDA. Also, the addition of TMPTA and EBI at 100 kGy improved all the physical properties, which provided suitable material based on natural polymer for biocomposites. The presence of TMPTA enhanced the crosslinking of LLDPE/CY during irradiation, which in turn enhanced the thermal stability of the composites. The natural soil burial and weathering of LLDPE/CY biocomposites were carried out for 1 year and the samples were collected and measured after 3, 6, and 12 months for degradability tests, by means of tensile tests, morphological study, and weight loss measurements. Within the 1-year exposure to soil burial and weathering test, the tensile strength and Eb of the composites decreased, while their Young's modulus increased. The deterioration in weight properties of the composites was investigated, where the presence of CY filler in the composites significantly accelerated the degradation of the LLDPE/CY biocomposites. The effects of cobalt stearate (CS) in natural soil burial and weathering on the properties of LLDPE/CY biocomposites were also examined, and the results indicate that the tensile strength and Eb of the composites decreased, while the Young's modulus increased with the increment in CY and CS contents. The morphology characterisation through SEM proved that the addition of CS to the composites accelerated their degradation.

CHAPTER 1: INTRODUCTION

1.1 Introduction

The awareness of environmental conservation and the use of green products have been emphasised in related industries for the past few decades. In response to this, numerous studies have been conducted, particularly in the polymer processing industry, to substitute the petroleum-based conventional plastic with plastic materials containing natural fibre. Natural fibre is produced from plants and animals which is also known as biofibre. Today, biofibre is widely used to reinforce various polymer matrix materials. It is widely used in research and industries because it offers various advantages over inorganic fillers such as cost saving, abundantly available, low density, and good deformability. The usage of biofibre is less abrasive to equipment and machinery during its processing. Biofibre is being extensively used for the production of cost-effective ecofriendly composites (Abdulsada & Al-Mosawi, 2015).

Biofibre is used in the processing of biocomposite using various types of polymeric matrix materials. The emergence of polymer matrix improves the interaction between filler and matrix, and leads to the enhancement of new methods or approaches (Saba et al., 2014). Biocomposites are used in several applications such as building construction, aerospace, automotive, and packaging industries.

Research and development of these bio-based materials will directly support our ecosystem and contributes to social and economic improvements in rural and remote areas in developing countries. Since biofibres are harmless compared to synthetic fibres, they could as well offer a resolution to the problem of environmental contamination with their new uses for waste materials. Cheap biofibres can be obtained in a plentiful amount in most developing countries, giving these countries a chance to use their own natural resources in their composite processing industry (Hassan et al., 2010). Biofibres was introduced as a filler in polymer blends as it is deemed beneficial to the environment, as far as the degradability and utilisation of renewable materials were concerned (Rout et al., 2001). Typical widely studied for their suitability as fillers in polymer blends include rice husks, flax fibres, and palm fibres.

In recent years, there has been a high consumption of polyethylene in the packaging and the agricultural sectors. Packaging waste has caused increasing environmental concerns (Vroman & Tighzert, 2009). The use of mulch films in agricultural sector, similarly, gives adverse environmental impacts with a high energy consumption required for the removal and disposal of their waste (Kyrikou & Briassoulis, 2007). This conventional plastic is a nondegradable plastic and its disposal in the natural environment can cause hazardous impact on the environment (Pavani & Rajeswari, 2014). Khabbaz and Albertsson (2001) reported that polyethylene has a very slow degradation rate of less than 0.5% in 100 years. The rate increases to 1%, if it is exposed to sunlight for 2 years.

The rapid development of biocomposites provides vast opportunities for people all over the world to improve their standard of living. Besides, it has been a great motivating factor for materials scientists to delve into the potentials provided by the composites. Since many of the renewable materials are based on agricultural products as a source of raw materials, particularly to plastic industries, a non-food source of economic development for farming and rural areas in developing countries could be generated. For example, generation of jobs by agro-based materials can be provided by the use of rice husk, which constitutes more than 10% of world rice production. It was reported that the increasing use of renewable materials would create or secure employment in rural areas, especially in sectors such as forestry and agriculture (Satyanarayana et al., 2009).

To overcome these problems, biodegradable plastics are produced from various renewable materials, or blends of synthetic materials and biofillers. Also, biofibres are readily available, cheap, and biodegradable, making them advantageous to be used as plastics (Iovino, Zullo, Rao, Cassar, & Gianfreda, 2008). Biofibres have been increasingly used as reinforcing materials for polymer-based matrices, not only because of their environmentally friendly nature, but also because of their low cost and intrinsically interesting properties such as low density, good shape ratio, and superior mechanical behaviour. Due to the increased awareness of ecological safety and utilisation of renewable materials towards a greener society, the use of natural fibres in the industries as biofiller or reinforcement materials in composites also increases by leaps and bounds (Satyanarayana, Arizaga, & Wypych, 2009).

In general, various types of fillers were incorporated into LLDPE matrix due to its physical properties and cost-saving feature. Organic fillers such as biofibres or powders derived from renewable natural resources such as banana, sisal, hemp, jute, pineapple, bamboo, cotton, coconut, rice husk, and kenaf have been used recently, due to their advantages in terms of high strength and stiffness, low cost, low density, low CO_2 emission, biodegradability and annually renewable (Ashori, A. 2008).

Biofibres displays comparatively poor fibre/matrix interactions, water resistance, and relatively lower durability. This consequently causes weaker interfacial or adhesion bonds between highly hydrophilic natural fibres and hydrophobic nonpolar organophilic polymer matrix, resulting in a significant impairment of the properties of the composites, thus depriving them of potential industrial use and output. To overcome this compatibility deficiency, a number of approaches have been established, such as the introduction of coupling agents and/or various surface modification techniques (Susheel Kalia, Kaith, & Kaur, 2009). Modification of the biofibres surface is achieved by the use of physical, mechanical, and/or chemical substance.

1.2 Problem Statements

Plastic waste is a hazardous environmental problem in Malaysia and polyethylene (PE) constitutes one of the greatest pieces of plastic waste. PE is currently in high demand for its use in various packaging applications. Although plastic recycling is a useful alternative to reduce plastic waste in the surroundings, its operations, sometimes, can be very costly, compared to the production of new plastic. Petroleum-based plastic is not biodegradable and it is inert to microorganisms, ultraviolet, heat, and water, thus causing disposal problems and jeopardising marine life. The most commonly employed method of municipal solid waste disposal is landfilling. Therefore, alternative materials, such as biodegradable plastic or biocomposite must be sought.