PREPARATION AND CHARACTERIZATION OF PINEAPPLE LEAF FIBER/POLY LACTIC ACID-BASED BIOCOMPOSITE

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By

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

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PREPARATION AND CHARACTERIZATION OF PINEAPPLE LEAF FIBER/POLY LACTIC ACID-BASED BIOCOMPOSITE

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April 2019

Chairman : Associate Professor Nor Azowa bt Ibrahim, PhD
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In this work, biocomposite from Polylactic acid (PLA) and Pineapple leaf fiber (PALF) was prepared with different fiber loadings 10-50 wt%. The composite was prepared via melt mixing process followed by hot press. Superheated steam (SHS) treatment at various temperature (190 - 230°C) and treatment time (30 - 120 min) have been applied on the fiber to enhance the surface adhesion between the polymer and the fiber. The best condition was at 220°C for 60 min as reflected by the tensile strength of the composite. Additionally, epoxidized palm oil (EPO) was added in the biocomposite as plasticizer at different percentages (1-5 wt%) to overcome the brittleness of the PLA and improve the performance of the biocomposite. A notable enhancement was observed in tensile strength with the addition of 1 wt% of the EPO.

The SHS treated fiber at 220°C for 60 min showed better characteristic compared to the untreated fiber. In term of chemical composition, the treated fiber showed a higher content of cellulose and lignin with lower content of hemicellulose. The FTIR results indicated the partial removal of the hemicellulose from the SHS treated fiber. Thermal stability of the SHS treated fiber was improved based on TGA thermogram. The XRD results showed higher crystallinity for the SHS treated fiber compared to the untreated fiber. Moreover, the SEM micrographs proved the removal of the impurities from the SHS treated fiber. It can conclude from these results that the SHS treatment successfully modified the chemical compositions and microstructure of fiber.

In term of fiber loading the highest tensile strength was obtained when 30 wt% of fiber loading was used. The results of the incorporation of the SHS treated fiber into the biocomposite showed the effectiveness of SHS treatment in increasing the PALF surface roughness due to the elimination of surface impurities and hemicellulose. The tensile, flexural and impact properties were notably improved by the presence of SHS-

PALF. Dimensional stability of the biocomposite showed a reduction in water uptake and thickness swelling of the biocomposite. additionally, the scanning electron microscopy analysis showed enhancement in interfacial adhesion between the PLA and the treated PALF.

In the case of incorporation of EPO into the biocomposite. The results showed that 1 wt% of EPO showed a significant improvement in the properties of the biocomposite. Tensile strength and tensile modulus as well as flexural and impact strengths were improved. Moreover, with increasing of the EPO content into the biocomposite it showed increment in the elongation at break. The scanning electron microscopy showed improvement in the interfacial adhesion as well with 1 wt% of EPO into the biocomposite.

In conclusion, this study proved the possibility of using agricultural waste crops to work as a filler into the polymer matrix, and SHS can be used to treat the fiber to improve the interfacial adhesion between fiber and matrix.

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

PENYEDIAAN DAN PENCIRIAN FIBER DAUN NANAS PENGUKUH POLI BIOKOMPOSIT

Oleh

AHMED JAAFAR HUSSEIN CHALLABI

April 2019

Pengerusi : Profesor Madya Nor Azowa bt Ibrahim, PhD
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Dalam kajian ini, biokomposit dari Asid Polilaktik (PLA) dan Fiber Daun Nanas (PALF) telah disediakan dengan nisbah fiber yang berbeza, iaitu 10-50 wt%. Komposit tersebut telah disediakan melalui proses percampuran lebur diikuti oleh penekan panas. Rawatan stim superpanas (SHS) pada pelbagai suhu (190 - 230°C) dan tempoh rawatan (30 - 120 min) telah diaplikasikan ke atas fiber bagi meningkatkan pelekatan permukaan antara polimer dan fiber. Keadaan yang terbaik adalah pada 220°C bagi 60 min seperti yang digambarkan oleh kekuatan tegangan komposit tersebut. Di samping itu, minyak kelapa sawit terepoksi (EPO) telah ditambahkan dalam biokomposit sebagai pemplastik pada peratusan yang berbeza (1-5 wt%) bagi mengatasi kerapuhan PLA dan memperbaiki prestasi biokomposit. Suatu peningkatan yang ketara telah dikesan dalam kekuatan tegangan dengan penambahan 1 wt% EPO.

Fiber terawat SHS pada 220°C bagi 60 min menunjukkan ciri yang lebih baik daripada fiber tanpa rawatan. Dari segi komposisi kimia, fiber terawat menunjukkan kandungan selulosa dan lignin yang lebih tinggi dengan kandungan hemiselulosa yang lebih rendah. Dapatan FTIR memperlihatkan penyingkiran hemiselulosa daripada fiber terawat SHS. Kestabilan termal fiber terawat SHS telah ditingkatkan berdasarkan termogram TGA. Dapatan XRD menunjukkan kehabluran yang lebih tinggi bagi fiber terawat SHS berbanding dengan fiber tanpa rawatan. Tambahan pula, mikrograf SEM membuktikan penyingkiran impuriti daripada fiber terawat SHS. Kajian ini menyimpulkan dari dapatan ialah rawatan SHS berjaya dalam pengubahsuaian komposisi kimia dan mikrostruktur fiber.

Dari segi pemuatan fiber, kekuatan tegangan tertinggi telah diperoleh ketika 30 wt% pemuatan fiber digunakan. Dapatan penggabungan fiber terawat SHS ke atas biokomposit menunjukkan keberkesanan rawatan SHS dalam meningkatkan

kekesatan permukaan PALF disebabkan penyisihan impuriti permukaan dan hemiselulosa. Sifat tegangan, lenturan dan impak terutamanya, telah meningkat dengan kehadiran SHS-PALF. Stabiliti dimensional biokomposit menunjukkan penurunan dalam penyerapan air dan ketebalan pembengkakan biokomposit. Tambahan pula, fiber terawat menunjukkan stabiliti termal yang lebih baik dari segi degradasi termal. Analisis mikroskopi elektron pengimbas menunjukkan penambahbaikan pelekatan antara muka antara PLA dan PALF terawat.

Kesan EPO sebagai pemplastik ke atas biokomposit telah dikaji dan peratusan pemplastik terbaik adalah pada 1 wt% EPO. Sekiranya biokomposit menunjukkan peningkatan yang signifikan dalam kekuatan tegangan dan modulus tegangan, didapati kekuatan lenturan dan impak juga meningkat. Di samping itu, pemanjangan pada tempoh rehat meningkat dengan peningkatan peratusan EPO ke dalam biokomposit. Mikroskopi elektron pengimbas menunjukkan peningkatan dalam pelekatan antara muka dengan penggabungan 1 wt% EPO ke dalam biokomposit.

Sebagai kesimpulan, kajian ini membuktikan kemungkinan menggunakan sisa tanaman pertanian yang berfungsi sebagai pengisi ke dalam matriks polimer dan SHS dapat digunakan bagi merawat fiber bagi memperbaiki pelekatan antara muka antara fiber dan matriks.

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

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LIST OF ABBREVIATIONS

ASTM	American Standard for Testing Methods
PALF	Pineapple leaf fiber
PLA	Polylactic acid
EPO	Epoxide palm oil
CrI	Crystallinity index
SHS	Superheated steam
FTIR	Fourier transform infrared
XRD	X-ray diffraction
SEM	Scanning electron microscopy
TGA	Thermogravimetric analysis
DTG	Differential thermogravimetric

CHAPTER 1

INTRODUCTION

1.1 Background of the study

Recently, there has been growing interest to produce biodegradable plastic products to replace the non-biodegradable plastics that used in most applications. The use of biodegradable plastics not only environmentally friendly, but also would control the pollution issues and the problems of waste disposal. Biodegradable plastics could be divided either synthetic or natural polymers. Natural polymers are available in large production and it comes from renewable resources, while synthetic polymers are produced from non-renewable petroleum resources.

Biodegradable plastics and polymers were first introduced in 1980s (Vroman and Tighzert, 2009). Biodegradable polymers can be divided into different categories based on the raw materials (renewable and non-renewable) and the process used in their production (Luckachan and Pillai, 2011). However, the production for these biodegradable polymers is relatively high in cost.

Several attempts have been done in order to reduce the cost of these biodegradable polymers in manufacturing, such as combining them with relatively low cost filler to produce cost-effective composite. One of the fillers that have been commonly used to reinforce these biodegradable polymers is the agricultural waste. It can produce a cost-effective bio-composite and can change the characteristics of the biodegradable polymers. Natural fibers such as flax, hemp, jute, oil palm and kenaf have drawn a remarkable attention in numerous applications for instance packaging, furniture, construction and automotive (Shalwan and Yousif, 2013). Further, the agricultural wastes were identified as a suitable for paper production, composite and engineered material (Dungani *et al.*, 2016).

In the past few years, lots of studies have reported the use of natural fiber waste as a reinforcement agent in polymer composites production. This is mainly due to the properties of natural fibers such as light weight, high strength, low cost, eco-friendly, renewable and their flexibility during processing. Most of these fibers give good mechanical performance and improved the properties for the polymer composite.

1.2 Problem statements

Agricultural wastes are increasing rapidly due to the increasing of production and the modern development of this sector, which as well representing threats to the environment. These wastes generated from different types of plant fibers such as jute,

sisal, pineapple, coir, banana, oil palm and coconut which all are abundantly available and annually renewable.

Malaysia is considered one of the important countries for pineapple production. According to the statistics showed by the Malaysian Pineapple Industry Board (MPIB) in 2016, Malaysia production for pineapple was 391,714 tonnes from 13,149 hectares harvested area and Johor was leading with the highest annual production of 273,949 tonnes. This large production of pineapple for Malaysia can generate more wastes, as it was recorded that the waste generated from this production annually in Johor was around 188 tonnes. Usually, these wastes will be burnt after three times the pineapple yielding, which can cause an environmentally issues and can affect the surrounding. Pineapple leaf fiber (PALF) is one of the waste materials in agriculture sector with very large production in Malaysia as well as Asia. In this work PALF was utilized by using them as a filler to reinforce thermoplastic biodegradable polymer to fabricate biocomposites. This could reduce the environmental problems caused by burning PALF. At the moment there are limited researches conducted on the use of Malaysian PALF wastes in composite fabrication. Thus, without doubt the use of PALF as a filler to produce biocomposites can add a value to the Malaysian PALF wastes in the future.

Natural fibers have showed excellent properties for biocomposites fabrication. However, natural fibers still have some drawback when it is used as a filler to reinforce polymer matrix for example, lack of good interfacial adhesion between the fiber and the polymer, low melting point, hydrophilic nature of the fiber that can cause high moisture absorption and swelling property which leads to creation of cracks in brittle matrices. Therefore, attempts have been done to overcome these drawbacks in the fibers, by modifying the surface of the plant fibers. It showed increment in surface roughness and decrement in moisture absorption. Various methods have been done in order to improve the compatibility between the hydrophilic fibers and hydrophobic polymer matrix. Most of treatment methods that have been used previously involved the use of chemicals for instance, alkali, silane, acetylation and benzoylation. These methods using chemicals, which could be not eco-friendly, toxic and expensive. Therefore, the efforts should be focused on environmentally friendly methods to increase the hydrophobicity of natural fibers. In this work superheated steam (SHS) was chosen to modify the surface of the fibers because SHS method is cheap and eco-friendly. According to our knowledge up to the present time, there is no work conducted on the utilization of SHS to modify PALF surface.

Polylactic acid (PLA) inherent some drawbacks such as high cost and brittleness. The attempts to overcome these issues were by blending PLA with natural fibers to reduce the cost and blend PLA with other polymers and modifying PLA with plasticizers. In this work epoxide palm oil (EPO) was used as plasticizer to modify the PLA and study its effect of the mechanical performance of the biocomposite.

1.3 Significance of the study

This work is focusing on producing environmentally friendly biocomposite material from biodegradable thermoplastic and natural fiber via melt blending technique followed by hot-press. The PALF was chosen in this work due to the fact that it is one of the Malaysian agricultural waste crops. Therefore, there is no doubt that using PALF from Malaysian cultivars can add a value to the pineapple production in Malaysia. In addition, the plasticizer used in this work is EPO due to its low cost, availability and non-toxic, and it is one of the vegetable oils. Finally, this work concerns on investigate the difference in the effect of fiber modification and effect of plasticizer on the biocomposite performance.

Objectives

The objectives of this study were:

1. To modify PALF surface by superheated steam treatment.
2. To determine the dimensional stability, mechanical, thermal and morphological properties of SHS-PALF/PLA and PALF/PLA biocomposites.
3. To evaluate the effect of EPO on the dimensional stability, mechanical, thermal and morphological properties of the untreated and treated PALF/PLA biocomposites.

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