UNIVERSITI PUTRA MALAYSIA

THE EFFECTS OF SURFACE TREATMENT AND PLASTICIZATION ON PROPERTIES OF KENAF FILLED POLY(LACTIC ACID) COMPOSITES

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By

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THE EFFECTS OF THE SURFACE TREATMENT AND PLASTICIZATION ON PROPERTIES OF KENAF-FILLED POLY(LACTIC ACID) COMPOSITES

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Chairman : Nor Azowa Ibrahim, PhD

Faculty/Institute : Institute of Tropical Forestry and Forest Product

The main objective of this study is to produce low cost environmental friendly materials (bio-composites) for food containers to replace the existing synthetic polymer materials such polyvinyl choride (PVC) , polypropylene (PP) and polyethylene terepthalate (PET), which are not green product and non-compostable materials. The main materials for this product are the poly(lactic acid) as a biodegradable polymer matrix and kenaf bast fibre (KBF) as a reinforcement. This research consists of four interconnected parts: the study of bio-composites materials on the effects of the fibre loadings, the modification on the fibre surfaces with sodium hydroxide (NaOH) in order to roughen the fibre surface, the modification of poly(lactic acid) with the plasticizers, and the enhancement on the polymer composite materials by combining the surface modification treated fibres with the plasticized poly(lactic acid) to have more green materials with good performances. Research and development on the poly(lactic acid) were conducted to modify the rigidity properties of the polymer itself, through the addition of triacetin and glycerol as plasticizers. These types of

plasticizer are functionally compatible with the poly(lactic acid). This research was also done on fibre reinforcement to synergistically use the short KBF which has been effectively surface treated in order to improve the fibre matrix adhesion in the resulting bio-composites materials.

The incorporation of KBF with PLA, with more than 30 wt% fibre loading, has been found to improve the tensile strength and modulus, whereas loading with higher percentage reduces the tensile and modulus value. Meanwhile, the thermal stability of the materials reveals that the PLA matrix and KBF have lower thermal stability as compared to the PLA/ KBF composites. The thermogravimetric analysis shows that 30 wt% of KBF loading has less amount of water content in fibres as compared to 50 wt% KBF loading, hence improves the strength of the composites. Water absorption analysis revealed that the water uptake increased with the increase of the KBF loading filled PLA matrix.

The surface modification treatment with 4% of NaOH on KBF enhanced the plasticized PLA/KBF tensile strength, flexural strength, and impact strength as compared to plasticized PLA with untreated KBF composite systems. Dynamic mechanical analysis showed that with treated fibres, the composite had an increment in the storage modulus that was attributed to the enhancement of the fibre matrix adhesion. Tg from the loss modulus results showed that the plasticized PLA with the treated KBF composites shifted to a higher temperature.

The addition of 5% triacetin and glycerol as plasticizers to modify the brittleness of PLA demonstrated that the plasticized PLA/KBF, with good mechanical and thermo-mechanical properties, have been developed. Triacetin improves the compatibility between the PLA

matrix and KBF, whereas this is *vice versa* for the glycerol. The tensile strength properties of the plasticized PLA/KBF composites materials were significantly higher than those plasticized PLA/KBF with glycerol. The scanning electron microscopy photograph of the PLA/KBF composites plasticized with triacetin indicated the extent of the fibre-matrix interface adhesion. Meanwhile, the dynamic mechanical analysis showed that the PLA/KBF composites with triacetin have higher storage modulus as compared to the PLA/KBF with glycerol which corresponds to higher tensile modulus. The addition of both plasticizers has also been found to lower the softening temperature of the composites.

The biodegradability test showed that the increase in the fibre content in the PLA produced a rapid decrease in the percentage of weight loss by bacteria and fungi during the soil burial process. The degradation time taken by the PLA/treated KBF with glycerol is longer than those of the plasticized PLA/treated KBF composites with triacetin. KBF filled with plasticized PLA bio-composites is environmental friendly and degradable, in addition to the fact that it can be considered as an alternative to conventional plastic materials such as polypropylene for packaging, food containers and disposable products.

Abstrak tesis yang dikemukakan kepada Senat Universiti Putra Malaysia sebagai memenuhi keperluan untuk ijazah Doktor Falsafah

KESAN RAWATAN PERMUKAAN DAN PEMPLASTIKKAN TERHADAP CIRI KENAF DIISI POLI(LAKTIK ASID)

Oleh

MAIZATULNISA BT OTHMAN

December 2010

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Objektif utama dalam kajian ini adalah untuk menyediakan bahan kos rendah yang mesra alam sekitar untuk bekas makanan bagi menggantikan polimer yang sedia ada seperti polivinil klorida (PVC), polipropilena (PP), dan polietilena tereptalat (PET) yang tidak mesra alam dan tidak terurai. Bahan utama bagi produk ini adalah polimer boleh urai iaitu poli(laktik asid) dan gentian kulit kenaf sebagai agen pengukuh. Kajian ini merangkumi empat bahagian yang berhubungkait: kajian ke atas bahan bio-komposit terhadap kesan penambahan gentian, modifikasi rawatan permukaan ke atas permukaan gentian menggunakan natrium hidroksida (NaOH) untuk permukaan gentian, modifikasi ke atas poli(laktik asid) menggunakan pemplastikan, dan memperkukuhkan bahan polimer komposit dengan menggabungkan modifikasi permukaan gentian terawat dengan poli(laktik asid) terplastikan bagi tujuan untuk menjadikannya lebih mesra alam di samping mempunyai ciri-ciri yang baik. Kajian dan pembangunan ke atas poli(laktik asid) telah dilakukan bagi mengubah suai sifat simpulan polimer tersebut dengan penambahan triasitin dan gliserol sebagai bahan pemplastikan.

Bahan pemplastikan ini mempunyai fungsi yang bersesuaian dengan poli(laktik asid). Kajian ini juga telah dilakukan ke atas memperkukuhkan gentian menggunakan gentian kulit kenaf yang telah menunjukkan keberkesanan rawatan permukaan untuk memperbaiki kesesuaian gentian dalam penghasilan bio-komposit.

Penggabungan antara KBF dengan PLA lebih daripada 30% penambahan gentian telah memperbaiki kekuatan dan modulus regangan, tetapi penambahan gentian lebih daripada peratusan ini telah mengurangkan nilai kekuatan dan modulus regangan. Kestabilan termal menunjukkan bahan PLA dan KBF mempunyai kestabilan termal yang rendah berbanding PLA/KBF komposit. Analisis TGA menunjukkan penambahan 30% KBF mempunyai kadar resapan lembapan yang rendah di antara gentian maka membantu perlekatan antara PLA dan kulit kenaf, seterusnya meningkatkan kekuatan tensil. Analisis serapan air menunjukkan kadar serapan meningkat dengan peningkatan penambahan KBF di dalam matrik PLA.

Rawatan permukaan dengan 4% NaOH ke atas KBF mengukuhkan lagi sifat kekuatan regangan, lenturan, dan hentaman PLA/KBF dengan pemplastikan berbanding PLA dengan pemplastikan dengan KBF tanpa rawatan. Keputusan dari analisa mekanik dinamik mendapati gentian dengan rawatan, komposit memberikan peningkatan terhadap storan modulus, menunjukkan pengukuhan antara matrik dan gentian. Nilai Tg daripada keputusan modulus kehilangan menunjukkan komposit PLA terliat dengan KBF dengan rawatan telah beralih kepada suhu yang lebih tinggi.

Penambahan 5% triasitin dan gliserol sebagai bahan pemplastikan untuk memperbaiki kerapuhan PLA menunjukkan bahawa PLA/KBF dengan pemplastikan yang mempunyai sifat mekanikal dan termal-mekanikal yang baik boleh dibangunkan. Triasitin memperbaiki kesesuaian antara PLA dan KBF manakala gliserol pula sebaliknya. Kekuatan regangan komposit PLA/KBF terplastik adalah lebih tinggi berbanding komposit PLA/KBF terplastikan dengan gliserol. Analisis SEM terhadap komposit PLA/KBF dengan pemplastikan triasitin mendapati mengukuhkan lekatan antara gentian dan juga polimer. Analisis DMA juga menunjukkan PLA/KBF dengan pemplastikan triasitin mempunyai storan modulus yang tinggi berbanding PLA/KBF dengan pemplastikan gliserol yang mempunyai regangan modulus yang tinggi. Penambahan bahan pemplastikan telah merendahkan suhu pelembutan komposit.

Ujian biodegradasi menunjukkan sampel yang mengandungi peratusan gentian yang tinggi sangat cepat terurai oleh bakteria dan fungi dan meningkatkan peratusan berat hilang dengan cepat. Tempoh biodegradasi yang diambil oleh PLA/ KBF terawat dan gliserol adalah lebih lama berbanding PLA/KBF terawat dan triasitin. Komposit PLA/KBF terplastikan adalah mesra alam dan bahan boleh urai yang boleh dipertimbangkan sebagai bahan alternatif kepada plastik seperti polipropilena dalam produk pembungkusan, bekas makanan dan produk pakai buang.

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I certify that an Examination Committee met on **24 December 2010** to conduct the final examination of **Maizatulnisa Bt Othman** on her Doctor of Philosophy thesis entitle "**The effects of surface treatment and plasticization on properties of kenaf filled poly(lactic acid) composites**" in accordance with Universiti Pertanian Malaysia (Higher Degree) Act 1980 and Universiti Pertanian Malaysia (Higher Degree) Regulations 1981. The committee recommends that the candidate be awarded the relevant degree.

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

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DECLARATION

I declare that the thesis is my original work except for quotations and citations, which have been duly acknowledged. I also declare that it has not been previously, and is not concurrently, submitted for any other degree at Universiti Putra Malaysia or at any other institutions.

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MAIZATULNISA BT OTHMAN

Date: 24 December 2010

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