UNIVERSITI PUTRA MALAYSIA

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By

INTAN SYAFINAZ MOHAMED AMIN TAWAKKAL

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

February 2011

DEDICATION

To my beloved mother, Rahimah Mahmud;

My loving brothers, Hafiz and Azhar;

My family members and friends.

Thanks for your encouragement, patience and loving support.

Abstract of thesis presented to the Senate of Universiti Putra Malaysia in fulfilment of the partial requirement for the degree of Master of Science

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Chair : Rosnita A. Talib, PhD

Faculty : Engineering

The aim of current research was to prepare kenaf derived cellulose (KDC)/polylactic acid (PLA) composites. The KDC or α-cellulose was derived from kenaf bast fibre (KBF) which was chemically treated via chlorination and mercerisation processes. The characteristics of kenaf fibre and cellulose on the chemical, morphological and physical properties were investigated by using FTIR, ESEM and density measurements. Optimisation of the composite processing variables namely temperature and time was carried out via RSM with respect to tensile strength and stock temperature. The composites were compounded using internal mixer at fixed KDC loading of 5 wt% and screw speed of 50 rpm in the temperature range of 160, 170 and 180°C for 10, 20 and 30 minutes. The effects of various KDC loadings (0-60 wt%) on the chemical, mechanical, thermal and physical properties of the KDC/PLA composites were also investigated.

The cellulose was successfully derived from the KBF by removing of lignin and hemicellulose. This was confirmed by the absence of these components from the FTIR spectrum of the cellulose and the ESEM micrographs. From the ESEM micrographs, the cellulose could be observed by its greater size reduction than the raw fibre and rough surface topography. Meanwhile, the ANOVA statistical data demonstrated that the tensile strength and stock temperature of the 5 wt% KDC/PLA composites were affected mostly by mixing temperature with $P < 0.0500$. The composites performed an optimum value at two combinations of variables which are at 170°C for 20 minutes and 170°C for 30 minutes. Nevertheless, 170°C and 30 minutes was chosen as the best mixing conditions in order to produce composite at higher KDC loading.

The FTIR spectrum of the KDC/PLA composite clearly indicated that no chemical interaction was present between KDC and PLA matrix in the composite. Interestingly, the incorporation of KDC into PLA matrix has demonstrated remarkable improvement in the tensile strength and stiffness properties. The tensile strength and modulus of KDC/PLA composite increased as the KDC loading increased. The ESEM micrographs revealed evidence of good interfacial adhesion between the KDC and PLA matrix and even distribution of the KDC in the composite system. However, the effects of KDC loading on the flexural and notched impact strength indicated that no remarkable changes were occurred in a range of 10 to 40 wt% of KDC loading.

The incorporation of the KDC into PLA matrix also improves the stiffness of composite due to the enhancement of storage modulus as compared to the neat PLA.

The DMA results demonstrated that the storage modulus of the 60 wt% KDC/PLA composite is twice higher than the neat PLA and the rest of the composites within a high temperature range (above 80°C). The glass transition temperatures (T_g) generated from the loss modulus curves exhibit that the peak was shifted to higher temperature as the percentage of cellulose loading in the composites was increased. Thus, the addition of KDC into the PLA matrix resulted better thermal stability and exhibited effective reinforcing agent. The water absorption of the composites increased with increasing KDC loading while the neat PLA absorbed lesser water. The 60 wt% KDC/PLA composite demonstrated approximately 12% of water uptake, which is considered as relatively low percentage of water absorption in biocomposite.

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

PENYEDIAAN KOMPOSIT SELULOSA TERBITAN DARIPADA KENAF TERISI POLILAKTIK ASID

Oleh

INTAN SYAFINAZ MOHAMED AMIN TAWAKKAL

Februari 2011

Pengerusi : Rosnita A. Talib, PhD

Fakulti : Kejuruteraan

Tujuan penyelidikan ini adalah untuk menghasilkan komposit selulosa terbitan kenaf (KDC) dengan polilaktik asid (PLA). KDC atau α-selulosa telah diterbitkan daripada gentian batang kenaf (KBF) melalui proses rawatan kimia iaitu pengklorinan dan pengalkalian. Ciri-ciri gentian kenaf dan selulosa terhadap sifat kimia, mofologikal dan fizikal telah dikaji dengan menggunakan teknik seperti FTIR, ESEM dan ukuran ketumpatan. Pengoptimuman pembolehubah-pembolehubah pemprosesan komposit terutamanya suhu dan masa telah dijalankan terhadap kekuatan regangan dan suhu terkumpul dengan menggunakan kaedah RSM pada peratusan kandungan KDC sebanyak 5 wt% dan kelajuan skrew sebanyak 50 rpm dengan menggunakan pengadun pada julat suhu 160, 170 dan 180°C untuk 10, 20 dan 30 minit. Kesan-kesan komposit bagi pelbagai peratusan kandungan KDC terhadap sifat kimia, mekanikal, termal dan fizikal turut dikaji.

Terbitan selulosa daripada KBF telah berjaya dihasilkan melalui penyingkiran lignin dan hemisellulosa. Penyingkiran ini disahkan dengan ketiadaan komponen-komponen tersebut di dalam spektrum FTIR selulosa dan mikrograf ESEM. Melalui mikrograf ESEM, didapati selulosa memiliki garis pusat yang lebih kecil berbandingan gentian kenaf dan topografi permukaan selulosa yang kasar telah diperolehi. Selain itu, berdasarkan data statistik ANOVA, didapati hubungan diantara kekuatan regangan dan suhu terkumpul terhadap pembolehubah komposit adalah amat dipergaruhi oleh suhu pengadun dengan $P < 0.0500$. Komposit-komposit ini juga menghasilkan nilai optimum pada dua kombinasi pembolehubah iaitu pada 170°C dan 20 minit serta 170°C dan 30 minit. Bagaimanapun, 170°C dan 30 minit adalah kombinasi pembolehubah yang paling sesuai bagi menghasilkan komposit pada peratusan kandungan KDC yang lebih tinggi.

Spektrum FTIR komposit secara jelas menunjukkan tiada interaksi kimia yang wujud diantara KDC dan matrik PLA. Menariknya, penggabungan KDC di dalam matrik PLA telah menunjukkan peningkatan kekuatan regangan dan ketegaran komposit yang memberangsangkan. Kekuatan dan modulus regangan juga meningkat dengan penambahan peratusan kandungan KDC. Mikrograf ESEM komposit pada permukaan patah kekuatan regangan membuktikan kewujudan lekatan yang baik diantara KDC dan matrik PLA serta taburan KDC yang sekata di dalam sistem komposit. Walau bagaimanapun, sifat komposit terhadap kekuatan lenturan dan impak menunjukkan tiada perubahan ketara berlaku pada peratusan kandungan KDC dari 10 hingga 40%.

Penggabungan KDC di dalam matrix PLA juga menambahkan ketegaran komposit. Ini disebabkan oleh peningkatan modulus simpanan komposit apabila dibandingkan dengan PLA tulen. Pada suhu yang tinggi (lebih 80°C), ujian DMA menunjukkan modulus simpanan komposit pada 60% kandungan KDC adalah dua kali lebih tinggi berbanding PLA dan komposit-komposit lain. Manakala, suhu peralihan kaca (T_g) yang diperoleh melalui lengkungan modulus lesapan menunjukkan berlakunya anjakan puncak ke suhu yang lebih tinggi dengan penambahan peratusan kandungan KDC. Oleh itu, penambahan KDC ke dalam matrix PLA telah menghasilkan kestabilan termal yang lebih baik serta membuktikan keberkesanan selulosa sebagai agen penguat. Penyerapan air terhadap komposit didapati semakin meningkat dengan penambahan peratusan kandungan KDC. Komposit yang mengandungi 60% kandungan KDC menunjukkan kadar penyerapan air sebanyak 12%. Peratusan ini bagaimanapun adalah agak rendah di dalam biokomposit.

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I certify that a Thesis Examination Committee has met on 24 February 2011 to conduct the final examination of Intan Syafinaz Mohamed Amin Tawakkal on her thesis entitle "Preparation of Kenaf Derived Cellulose-Filled Polylactic Acid Composites" in accordance with the Universities and University Colleges Act 1971 and the Constitution of the Universiti Putra Malaysia [P.U.(A) 106] 15 March 1998. The Committee recommends that the student be awarded the Master of Science.

Members of the Thesis Examination Committee were as follows:

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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 **Master of Science.** 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 institution.

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INTAN SYAFINAZ MOHAMED AMIN TAWAKKAL

Date: 24 February 2011

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