UNIVERSITI PUTRA MALAYSIA

IMPREGNATION OF IN-HOUSE SYNTHESIZED CARBON NANOTUBES INBACTERIAL CELLULOSE

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By

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

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IMPREGNATION OF IN-HOUSE SYNTHESIZED CARBON NANOTUBES IN BACTERIAL CELLULOSE

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Chair: Dayang Radiah Bt. Awang Biak, PhD

Faculty: Faculty of Engineering

Bacterial cellulose (BC) synthesized by *Acetobacter xylinum* has been discovered as a potential matrix for impregnating carbon nanotubes (CNT). The main objective of this study is to produce bacterial cellulose/carbon nanotube (BC/CNT) with improved hydrophobic surface. CNT addition is by mean to improve the electrical property of BC to be a conductive membrane. Hydrophobic membrane is more practical in device development since it can adhere with other hydrophobic material, which is achieved in this study by acetylation process. In this work, the membranes were produced using date extract (DE) culture medium. Three CNT impregnation techniques were tested, *i.e.* immersion, dispersion and spraying, whilst the acetylation conducted were homogeneous and heterogeneous processes.

In this study, morphological characteristic is an important property for sample selection to be characterized further. BC/CNT nanocomposite produced via spraying technique has uniform CNT dispersion and has the most dielectric conductivity value *i.e.* ~4.66 x 10^{-1} S/cm than the other techniques. Therefore, spraying was used to prepare BC/CNT acetate nanocomposite. Since the morphology of BC/CNT acetate obtained from heterogeneous process proves that the acetylation only occurred on the surface and the inner core structures of the BC are still intact, this process was used further.

The most significant characteristic of BC/CNT acetate is the existence of C=O, CH_3 and C-O functional groups of acetyl and higher water contact angle (~77°) than that of BC/CNT (~45°). When tested, decomposition and glass transition temperatures of BC/CNT acetate have reduced by 203°C and 35°C respectively, compared to that BC/CNT. The reduction is occurred mainly because the swollen BC structure of BC/CNT acetate is easily degraded by heat treatment.

The acetylated BC/CNT on the other hand, has higher crystal BC fraction, *i.e.* 51% compared to that of BC/CNT (49%). These findings indicate that the CNT hinders the degradation of nanocomposite and protects the BC crystals. Thus when acetylated, both elasticity and crystallite size of BC/CNT has only reduced by 9%, *i.e.*, from 34 to 31 GPa and from 27.3 to 24.9 nm, respectively. The bigger pore size of the acetate nanocomposite (~22 Å) than that of BC/CNT (~-103 Å) might allows further functionalization process introduced to the membrane.

As a conclusion, the CNT impregnation by spraying technique during the growing fermentation of BC coupled with heterogeneous acetylation treatment resulted in a more water resistant,

increased dielectric conductivity and increased crystal BC fraction. While, the sheet elasticity, crystallite size, surface area, decomposition and glass transition temperatures became lower. These characteristics makes acetylated BC/CNT sheet a potential as a biomaterial membrane in device development.

Abstrak tesis yang dikemukan kepada Senat Universiti Putra Malaysia sebagai memenuhi keperluan untuk Ijazah Master Sains.

IMPREGNASI NANOTIUB KARBON YANG DISINTESIS SECARA DALAMAN KE DALAM SELULOSA BAKTERIA

Oleh

SHARMEEN BINTI ADNAN

Ogos 2011

Pengerusi: Dayang Radiah Bt. Awang Biak, PhD

Fakulti: Fakulti Kejuruteraan

Selulosa bakteria (BC) yang disintesis oleh *Acetobacter xylinum* telah dikenalpasti sebagai matriks yang berpotensi tinggi untuk diisi dengan nanotiub karbon (CNT). Objektif utama penyelidikan ini adalah untuk menghasilkan nano-komposit selulosa bakteria/nanotiub karbon (BC/CNT) dengan permukaan hidrofobik. CNT ditambahkan bagi memperbaiki ciri elektrikal BC seterusnya menghasilkan membran yang konduktif. Membran yang hidrofobik pula lebih praktikal dalam penghasilan peralatan kerana ia lebih sesuai untuk digunakan bersama bahan hidrofobik yang lain. Dalam penyelidikan ini, membran dihasilkan menggunakan media kultur daripada ekstrak kurma. Tiga teknik impregnasi CNT yang dijalankan adalah rendaman,

serakan dan semburan manakala proses asetilasi pula adalah proses asitilasi penuh dan asitilasi separa.

Dalam kajian ini, morfologi adalah ciri penting dalam pemilihan sampel yang akan dianalisa sepenuhnya. Nano-komposit BC/CNT yang dihasilkan melalui teknik semburan menunjukkan keseragaman penyebaran CNT serta mempunyai nilai konduktiviti di-elektrik yang paling baik iaitu ~4.66x 10^{-1} S/cm berbanding nilai yang ditunjukkan oleh BC/CNT yang dihasilkan melalui teknik-teknik yang lain. Maka, teknik semburan telah digunakan untuk menghasilkan nano-komposit BC/CNT asetat. Disebabkan morfologi BC/CNT asetat yang disediakan melalui proses asitilasi separa menunjukkan degradasi hanya berlaku di permukaan sahaja sementara struktur teras dalam BC tidak terjejas, maka proses ini digunakan untuk penghasilan sampel seterusnya.

Ciri BC/CNT asetat dikenalpasti melalui kewujudan ikatan-ikatan yang mewakili kumpulan berfungsi asetil iaitu C=O, CH_3 dan C-O dan juga nilai sudut antara air dan BC/CNT asetat yang lebih tinggi (~77°) berbanding BC/CNT (~45°). Apabila kedua-duanya dikaji, suhu penguraian dan peralihan kaca bagi BC/CNT asetat menurun kepada 203°C dan 35°C, masing-masing. Penurunan ini berlaku kerana struktur BC dalam BC/CNT asetat yang membengkak memudahkannya diuraikan dengan rawatan suhu.

Namun begitu, BC/CNT asetat mempunyai kandungan hablur BC yang lebih tinggi, iaitu 51% berbanding BC/CNT (49%). Ini menunjukkan CNT telah menghalang proses penguraian nano-komposit seterusnya melindungi hablur BC. Disebabkan itu, kedua-dua keupayaan elastik dan saiz hablur BC/CNT hanya menurun sebanyak 9% sahaja, iaitu daripada 34 ke 31 GPa dan daripada 27.3 ke 24.9 nm, masing-masing. Tambahan pula, liang nano-komposit asetat yang lebih besar (~22 Å) berbanding BC/CNT (~-103 Å) mungkin membolehkan proses penambahbaikan fungsi dilakukan ke atas membran tersebut.

Kesimpulannya, impregnasi CNT melalui teknik semburan semasa fermentasi BC berlaku disamping rawatan asetilasi separa telah menghasilkan membran yang rintang kepada air, tinggi nilai konduktiviti dielektrik dan tinggi kandungan hablur BCnya. Namun, keupayaan elastik, saiz hablur, suhu penguraian dan suhu peralihan kaca telah menurun. Ciri-ciri ini menjadikan membran BC/CNT asetat berpotensi sebagai biomaterial membran dalam pembuatan peralatan.

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I certify that a Thesis Examination Committee has met on 25 August 2011 to conduct the final examination of Sharmeen Binti Adnan on her thesis entitled "Impregnation of In-House Synthesized Carbon Nanotubes in Bacterial Cellulose" 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 degree of Master of Science.

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This thesis was submitted to the Senate of Universiti Putra Malaysia and has been accepted as fulfillment 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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SHARMEEN BINTI ADNAN

Date: 25 August 2011

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