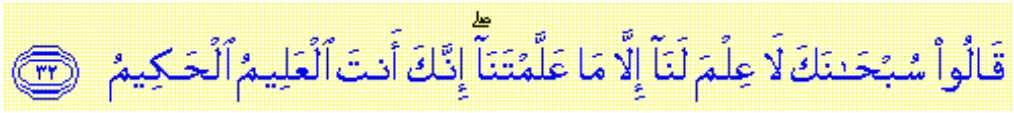
PREPARATION AND CHARACTERIZATION OF BIODEGRADABLE FILMS FROM SAGO STARCH AND POLY(VINYL ALCOHOL) BLENDS

By

SONNY WIDIARTO

**Thesis Submitted to the School of Graduate Studies,
Universiti Putra Malaysia in Fulfilment of the Requirement
for Degree of Master of Science**

April 2004



قَالُوا۟ سُبْحَٰنَكَ لَا عِلْمَ لَنَآ إِلَّا مَا عَلَّمْتَنَآ ۖ إِنَّكَ أَنتَ ٱلْعَلِيمُ ٱلْحَكِيمُ ﴿٣٢﴾

Surah Al Baqarah

Especially dedicated

to my beloved parents, Wiyatsari and Ir. Soedarsono S.

to my beloved wife, Renta Triantina, whose patience, supports and companionship has facilitated my work.

to my children, Abdullah al Mubarok, Abdurrahman Ahmad and Azkia Hanifa

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

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Chairman: **Associate Professor Mohamad Zaki Ab. Rahman, Ph.D**

Faculty: **Science and Environmental Studies**

Biodegradable films of sago starch and poly(vinyl alcohol) (PVA) blends (SP) have been prepared by solution casting method. Glycerol was used as a plasticizer. Sodium borate (borax) and glutaraldehyde were used to increase the compatibility of sago starch and PVA. The optimum amount of glycerol is 20% (w/w). The optimum values of tensile strength and elongation at break of the films are obtained with an addition of 8% (w/w) sodium borate (borax) or 62.5% (w/w) glutaraldehyde.

Both pure PVA and sago starch films are apparently transparent. However, their blend films are slightly opaque probably due to phase separation. Increase the amount of PVA in the film leads to an increase of the tensile strength and reduces the elongation of films. Both borax and glutaraldehyde increase the elongation at break of the sago starch – PVA blend films, but improvement of tensile strength is only obtained when the films are added with borax.

The films produced were characterized by FTIR spectroscopy, differential scanning calorimetry (DSC), thermogravimetry analysis (TGA), X-ray diffraction (XRD), and scanning electron microscopy (SEM). FTIR spectrum of sago starch – PVA – borax (SPB) film showed the shifting of characteristic O-H stretching vibration peak to a higher wavelength number at 3426 cm^{-1} and an increase of O-H bending intensity of absorbed water peak at 1638 cm^{-1}, indicating more tightly bound water present in the film in comparison with that of SP film. SEM studies show that SPB film was porous. Meanwhile, TGA studies revealed that decomposition of SPB film is a three-stage process. XRD studies indicated that the degree of crystallinity of SPB film is higher than that of SP film.

FTIR spectrum of sago starch – PVA – glutaraldehyde (SPG) film shows the appearance of aldehyde's carbonyl group at 1717 cm^{-1}. This peak should be disappeared after the acetalation reaction. However, the peak is weaker than that of normal aldehyde, indicating the occurrence of incomplete acetalation reaction. SPG film has a more compact structure than that of SP film as shown by SEM. Thermal decomposition of SPG film is also in a three-stage process. XRD studies indicated that the degree of crystallinity of SPG film is lower than that in SP film.

Tensile strength of both SPB and SPG films increase after storage treatment, whereas their elongation decrease. The changes of tensile properties could be related to both the changes on crystallinity and the loss of plasticizer during storage. The B-type crystallinity appeared on the SPB samples after two months of storage, but the same crystallinity was not observed in SPG samples.

Biodegradation of original and modified sago starch – PVA blend films have been studied by soil burial method. Percentage of weight loss of both SPB and SPG films increase with an increase of sago starch content.

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

PENYEDIAAN DAN PENCIRIAN FILEM BIO-URAI DARIPADA CAMPURAN KANJI SAGU DAN POLI(VINIL ALKOHOL)

Oleh

SONNY WIDIARTO

April 2004

Pengerusi: **Profesor Madya Mohamad Zaki Ab. Rahman, Ph.D**

Fakulti: **Sains dan Pengajian Alam Sekitar**

Filem bio-urai daripada campuran kanji sagu dan poli(vinil alkohol) (PVA) disiapkan dengan menggunakan kaedah kasting larutan. Gliserol digunakan sebagai bahan pemplastik. Sodium borat (boraks) dan glutaraldehid digunakan untuk meningkatkan kesesuaian kanji dan PVA. Keadaan optimum yang diperolehi untuk gliserol adalah 20% (w/w). Nilai kekuatan tensil dan elongasi optimum diperolehi pada 8% (w/w) sodium borat atau 62.5% (w/w) glutaraldehid.

Filem daripada PVA mahupun kanji sagu adalah lutsinar. Bagaimanapun, filem yang diperoleh daripada campuran keduanya adalah sedikit legap, dimungkinkan akibat daripada pemisahan fasa. Peningkatan jumlah PVA di dalam filem menyebabkan peningkatan kekuatan tensil dan pengurangan elongasi. Boraks dan glutaraldehid telah meningkatkan elongasi pada filem kanji sagu – PVA, namun peningkatan pada kekuatan tensil diperolehi hanya selepas penambahan dengan boraks.

Filem yang dihasilkan telah dicirikan melalui spektroskopi infra merah transformasi Fourier, kalorimetri pengimbasan perbezaan, analisis termogravimetri, pembelauan sinar-X dan mikroskopi pengimbasan elektron. Spektrum infra merah bagi filem campuran kanji sagu – PVA – boraks (SPB) menampakkan anjakan puncak khas O-H ke bilangan gelombang yang lebih tinggi pada 3426 cm^{-1} dan peningkatan puncak serapan O-H pada 1638 cm^{-1} yang menandakan lebih banyak air terikat pada filem berbanding pada filem SP. Kajian mikroskop pengimbasan elektron menunjukkan struktur berliang, manakala kajian termogravimetri menampakkan adanya tiga tahap penguraian terma pada filem. Kajian pembelauan sinar-X menunjukkan darjah penghabluran bagi filem SPB lebih tinggi berbanding filem SP.

Spektrum infra merah bagi filem campuran kanji sagu – PVA – glutaraldehid (SPG) menampakkan serapan karbonil bagi aldehid pada 1717 cm^{-1}. Puncak ini semestinya hilang selepas tindak balas asetalasi. Namun demikian puncak yang dihasilkan lebih lemah berbanding dengan puncak aldehid biasa. Hal ini menandakan berlakunya tindak balas antara kumpulan hidoksil dan karbonil yang tidak sempurna. Kajian mikroskop pengimbasan elektron menampakkan struktur yang lebih padat pada filem SPG berbanding pada filem SP. Kajian termogravimetri menampakkan adanya tiga tahap penguraian terma pada filem SPG. Kajian pembelauan sinar-X menunjukkan darjah penghabluran bagi SPG lebih rendah berbanding SP.

Filem SPB dan SPG menampakkan peningkatan pada kekuatan tensil dan pengurangan pada elongasi yang nyata selepas penyimpanan. Perubahan tersebut mungkin disebabkan oleh perubahan pada penghabluran filem mahupun berkurangnya bahan pemplastik

selama proses penyimpanan. Penghabluran jenis B diperhatikan pada filem SPB selepas penyimpanan, namun tidak diperhatikan pada filem SPG.

Kajian pembio-uraian filem dibuat dengan menggunakan kaedah penanaman dalam tanah. Filem SPB dan SPG menampakkan peningkatan pada peratus kehilangan berat dengan meningkatnya kandungan kanji dalam filem.

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I certify that an examination committee met on 30 October 2004 to conduct the final examination of Sonny Widiarto on his Master thesis entitled "PREPARATION AND CHARACTERIZATION OF BIODEGRADABLE FILMS FROM SAGO STARCH AND POLY(VINYL ALCOHOL) BLENDS" in accordance with Universiti Pertanian Malaysia (Higher Degree) Act 1980 and Universiti Putra Malaysia (Higher Degree) Regulations 1981. The Committee recommends that the candidate be awarded the relevant degree. Members of the Examination Committee are as follows:

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

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Date:

DECLARATION

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

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SONNY WIDIARTO

Date:

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