

UNIVERSITI PUTRA MALAYSIA

INFLUENCE OF GAMMA RADIATION ON OPTICAL AND DIELECTRIC PROPERTIES OF DYED POLYVINYL ALCOHOL FILM DOSIMETERS

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By

AJIS BIN LEPIT

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

February 2004



Dedication

بسم الله الرحمن الرحيم

"Proclaim! (or Read!) In the name of thy Lord and Cherisher, Who created, # Created man, out of a (mere) clot of congealed blood: # Proclaim! and thy Lord is Most Bountiful, # He Who taught (the use of) the Pen, # Taught man that which he knew not".

Versus Al-alaq: 1-5

To my family

Zuraida Adam
Muhammad Akmal Zulhilmi
Muhammad Akram Irfan
Fatin Najihah

Who has given all the patience, encouragement, love and support.



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

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February 2004

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The influence of y-rays on the optical absorption and inelastic scattering, dielectric properties and conductivity of radiation-sensitive dyed polyvinyl alcohol (TB/PVA) film dosimeters containing chloral hydrate and acid-sensitive Thymol blue dye were evaluated for possible use as food irradiation indicators. The dyed PVA films of different concentrations of chloral hydrate were irradiated with the absorbed doses ranging up to 12 kGy using γ-rays from Cobalt-60. The dehydrochlorination of chloral hydrate and radiolysis of water molecules induced by ionising radiations accelerated the formation of hydrochloric acid in the polymer matrix, which caused the change in colour of the dosimeters from yellow to red at the critical doses depending on the concentration of chloral hydrate. This radiation-induced colour change was analysed using UV-Vis spectrometer, where the absorption spectra produced two visible maximal bands, peaking at 445 nm and 554 nm. The dose response at 445 nm and 554 nm increases and decreases respectively with absorbed dose. The inelastic Raman scattering spectra of photons corresponding to the Raman frequency shifts of unirradiated and irradiated films were measured using a dispersive Raman spectrometer, which provide direct evidence of molecular



structure changes induced by ionising radiation and the subsequent chemical effects. The spectral intensities of Raman shifts at 815, 1984, 2350 and 2560 cm⁻¹ bands correspond to C-Cl, C=O, C=C and S-H bonds respectively were studied, which provide the dose response to the molecular vibration of the dosimeters. From dielectric and conductivity studies it is found that the dyed polymer dosimeters are ionic polymer materials. The dielectric constant (ϵ'), dielectric loss (ϵ'') and the electrical conductivity $\sigma(\omega)$ characteristics of the dosimeters were measured at different frequencies ranging from 20 Hz to 1 MHz. The dielectric constant and dielectric loss increase with absorbed dose at low frequencies and are independent of dose at higher frequencies for all chloral hydrate concentration. The AC conductivity (σ) increases with absorbed dose and frequency due to the formation of radiationinduced free radicals, cations and anions in the polymer matrix and due to ejected electrons in the conduction bands. Thus, the resistance derived from the impedance measurement, decreases with absorbed dose. Finally, the films were subjected to stability tests using digital densitometry method at different time intervals during post-irradiation storage. The results show the change in optical density is minimal over the period of 70 days for all irradiated samples. This suggests the dosimeters have optical absorption stability characteristics for use as alternative radiationsensitive dosimeters in irradiation facilities as long as they are shielded from sunlight or fluorescent lighting by wrapping with black plastic bag.



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

PENGARUH SINAR GAMMA KEATAS SIFAT-SIFAT OPTIK DAN DIELEKTRIK METERDOS FILEM WARNA POLYVINYL ALCOHOL

Oleh

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Pengaruh sinar-y ke atas penyerapan optikal dan serakan tidak elastik, sifat-sifat dielektrik dan konduktiviti bagi sinar-sensitif meterdos warna filem polyvinyl alcohol (TB/PVA) yang mengandungi kloral hidrat dan asid sensitif Thymol biru telah dikaji untuk aplikasi kemungkinan sebagai indikator penyinaran makanan. Filem-filem yang berbeza kepekatan bahan kloral hidrat disinarkan dengan dos-dos kobalt-60. sehingga 12 kGv menggunakan sinar-γ dari penyerapan Penyahidroklorinan kloral hidrat dan radiolisis molekul air diaruhkan oleh sinaran sebagai pemangkin melalui pembentukan asid hidroklorik dalam bahan polimer, yang menyebabkan meterdos berubah warna dari kuning ke merah pada dos kritikal yang bergantung kepada kepekatan bahan kloral hidrat. Perubahan warna ini telah dianalisa menggunakan spektrometer UV-Vis, dimana penyerapan spektrum menghasilkan dua jalur maksima dalam julat cahaya-nampak pada 445 nm dan 554 nm. Dos tindakbalas pada 445 nm meningkat dan 554 nm menurun dengan kenaikan dos penyerapan. Serakan foton tidak elastik spektra Raman bagi filem bergantung kepada perubahan frekuensi Raman sebelum dan selepas penyinaran diukur menggunakan penyebaran spektrometer Raman, bagi menyediakan bukti secara terus



perubahan struktur molekul disebabkan oleh sinar mengion dan seterusnya kesan kimia. Keamatan spektra Raman pada jalur 815, 1984, 2350 dan 2560 cm⁻¹ adalah sejajar ikatan dengan C-Cl, C=O, C=C dan S-H adalah telah dikaji, disebabkan tindakbalas dos getaran molekul-molekul meterdos. Dari kajian dielektrik dan konduktiviti didapati bahan meterdos polimer warna adalah bahan-bahan polimer ion. Pemalar dielektrik (ϵ'), kehilangan dielektrik (ϵ'') dan konduktiviti elektrik $\sigma(\omega)$ bagi meterdos, diukur julat frekuensi yang berbeza dari 20 Hz hingga 1 MHz. Pemalar dielektrik dan kehilangan dielektrik bertambah dengan dos penyerapan pada frekuensi rendah dan tidak bergantung pada dos ketika frekuensi tinggi bagi semua kepekatan kloral hidrat. Konduktiviti a.u (σ) bertambah dengan dos penyerapan dan frekuensi dihasilkan oleh pembentukan radikal bebas sinar-teraruh, kation dan anion di dalam matrik polimer yang disebabkan elektron disuntik keluar dalam jalur konduksi. Oleh itu rintangan diperolehi dari pengukuran impedan, menurun dengan dos serapan. Akhir sekali, sampel filem dikehendaki untuk ujian kestabilan menggunakan digital meterketumpatan pada tempoh masa yang berbeza semasa simpanan selepas disinarkan. Keputusan menunjukkan bahawa perubahan ketumpatan optik adalah rendah sepanjang masa 70 hari disimpan untuk semua bahan yang telah disinarkan. Ini boleh dicadangkan meterdos itu mempunyai ciri-ciri kestabilan optik untuk digunakan sebagai meterdos sinar-sensitif alternatif dalam prasarana penyinaran selagi ianya disimpan dan dilindungi daripada cahaya matahari atau flouresen dengan membungkus dengan plastik berwarna hitam.



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I certify that an Examination Committee met on 19th February 2004 to conduct the final examination of Ajis Bin Lepit on his Master of Science thesis entitled "Influence of Gamma Radiation on Optical and Dielectric Properties of Dyed Polyvinyl Alcohol Film Dosimeter" 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. Members of the Examination Committee are as follows:

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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 of concurrently submitted for any other degree at UPM or other institutions.

AJIS BIN LEPIT

Date: 14.06.04

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impedance for all frequencies for PVA films doped with different



LIST OF ABBREVIATIONS/SYMBOLS

 γ -ray - Gamma Ray

BPB - Bromophenol Blue

CR - Cresol Red

DCP - 2,6 dichloro phenol indophenol sodium salt

PVA - Polyvinyl alcohol
PVB - Polyvinyl butyral
PVC - Polyvinyl chloride

PS - Polystyrene

PMMA - Polymethyl methacrylate

TB - Thymol blue

 ϵ' - Dielectric Constant ϵ'' - Dielectric Loss

tan δ - Dielectric Loss Tangent

τ - Time constant or relaxation time

σ - Conductivity

WHO - World Health Organization

MINT - Malaysia Institute for Nuclear Technology Research

IAEA - International Atomic Energy Agency
FAO - Food & Agriculture Organization

UV-Vis - Ultraviolet – Visible GIP - Good Irradiation Practice

kGy - kiloGray IR - Infrared

NaOH - Natrium hydroxide



GLOSSARY

This glossary to define a few words in common use in this thesis. Many other quantities and term are defined in appropriate locations in the text.

Absorbed dose Amount of energy deposited by ionizing radiation in a material per unit mass of the material. Usually expressed in the special radiological unit rad or in the SI unit Gray.

Anti-Stokes Raman Scattering Light scattering in which the photons gain energy as a result of photon-molecule collisions.

Chromophore Molecule or part of the molecule that absorbed light.

Dielectric Dielectric is a material in which energy can be stored by the polarization of the molecules. It is a material that increases the capacitance or charge storage ability of a capacitor. Ideally it is a non-conductor of electrical charge so that an applied field does not cause a flow of charge but instead a relative displacement of opposite bound charges and hence polarization of the medium.

Dipolar (orientational) polarization arises when randomly oriented polar molecules in a dielectric are rotated and aligned by the application of a field so as to give rise to a net average dipole moment per molecule. In the absence of the field the dipoles (polar molecules) are randomly oriented and there is no average dipole moment per molecule. In the presence of the field the dipoles rotated, some partially and some fully, to align with the field and hence give rise to net dipole moment per molecule.

Dose (D) Used broadly for energy deposited in matter from radiation. Used in dosimetry for the energy absorbed per unit mass of material, usually by ionization processes. Units are the rad and the Gray (Gy), which are equivalent, respectively, to ergs/g and 1 J/Kg. There, 1 rad = 1/100 Gray or cGy.

Dosimetry The calculation, measurements and other activities required for determining the radiation dose to be delivered.

Electronic polarization Electronic polarization is the displacement of the electron cloud of an atom with respect to the positive nucleus. Its contribution to the relative permittivity of a solid is usually small.

Excitation The addition of energy to a system, transferring it from its ground state to an excited state. Excitation of a nucleus, an atom, or a molecule can result from absorption of photons or from inelastic collision with other particles.

Free Radical A highly reactive chemical species carrying no charge and having a single unpaired electron in an orbital.

