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The use of positron annihilation Doppler broadening spectroscopy in the characterization of radiochromic dosimetry films

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HIGHLIGHTS

- The structural changes of two types of γ-irradiated radiochromic films were investigated using the PADB spectroscopy.
- Correlation between DB S-parameter and OD of films was observed.
- Dependence of DB parameters on the polymerization degree of films' active layer was discussed.
- The capability of PADB technique to detect the structure transition of films' active layer was demonstrated.

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ABSTRACT

The positron annihilation Doppler broadening (PADB) spectroscopy has been employed to probe the defects and structural changes of various types of materials. In this work, an investigation was carried out for the first time to use PADB spectroscopy in probing the γ irradiated radiochromic films. The Gaf-Chromic MD−55 and HD−V2 radiochromic films irradiated with absorbed doses ranges of 0−80 and 0−2000 Gy, respectively, were subjected to Doppler broadening measurements employing a HPGe γ−ray spectrometer. The Doppler broadening line−shape parameters (S and W) of 511 keV annihilation radiations were discussed in terms of the polymerization degree of the active components of these dosimetry films. The S- and W-parameters were found to be dependent on the active component structures of both films. A reasonable correlation was also found between the values of the S−parameter and the optical density of these radiochromic films. In addition, the Doppler broadening line−shape parameters were successfully used to provide explanation of the observed nonlinearity of these films at the end of their dynamic dose ranges. The results demonstrate the applicability of the PADB technique to probe the physical and chemical changes occurred in the active layer of the studied radiochromic films during the solid−state polymerization reaction caused by γ irradiation of different absorbed doses.

1. Introduction

Radiochromic films are used in radiotherapy as a dosimeter for quite some time. Their applications have been expanded and covered environmental, industrial and other nonclinical applications. Only the absorption of radiation causes “radiochromic reaction” which consequently leads to direct coloration of a media. Unlike other methods of measuring radiation doses, these films are self-developing in response to radiation and do not require any chemical or physical (e.g. thermal or optical) processes for development. The radiochromic reaction is defined as a solid−state polymerization that changes the film color to deep blue. The resulting color of the active layer of the film quantifies the absorbed dose. This change of color indicates that the active layer has undergone significant chemical modifications under exposure to radiation (AAPM, 1998; Butson et al., 2003; Wong, 2009).

The radiochromic films types MD−55 and HD-V2 were used for absorbed dose measurements for high-energy photons. The MD-55 film is comprised of two active layers on polyester substrates with total thickness of ~160 μm laminated by two adhesive tapes with total thickness of ~50 μm. The HD-V2 film is comprised of one

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active layer coated on a clear, 97 μm polyester substrate. The percentage values of the active layer thickness to the substrate thickness for MD-55and HD-V2 films are ~20% (or ~15.2% by adding the thickness of adhesive tapes) and ~8.2, respectively. The active layers of the MD−55 and HD-V2 are about 32 and 8 μm, respectively (Gafchromic® MD-55; Gafchromic® HD-V2). The response of both films are energy-independent for photons. The active component of MD−55 films consists of a double-substituted diacetylene monomer with one polar end, organized into a crystal (Rink, 2008). The response dose range of the MD−55 film is a linear up to ~70 Gy (Gafchromic® MD-55). The HD-V2 film is composed of polycrystalline diacetylene active layer, suspended in a synthetic or natural polymer. This film is also composed of a yellow marker dye incorporated into the active layer (Gafchromic® HD-V2; Ettlinger et al., 2012). The dynamic dose range of the HD-V2 film is from 10 to 1000 Gy.

Positron annihilation spectroscopy (PAS) is a powerful tool that have been used to investigate the structural changes of various types of materials. Positron lifetime, Doppler broadening and angular correlation are the three main techniques of PAS can be used to extract basic structural features of studied samples (Schrador and Jean, 1988; Dupasquier and Mills, 1995). The PAS has been used to probe the defects and open-volumes in different types of solids (Marques-Netto et al., 2001; Abdel-Hady and Mohamed, 2002; Ferreira Marques et al., 2003; He et al., 2004; Abdel-Rahman et al., 2006a; Flounis-Mokrani, 2013). The embedded positron can preferentially be localized at various types of defects in solids and annihilate with an electron of the atoms of materials through γ-ray annihilation radiation. Useful information about the electronic environment around the annihilation site can be obtained with these γ rays. In polymers, many of incident positrons annihilate through a positronium (Ps), the bound state of a positron and an electron. The use of positrons as a probe to study the polymeric materials is preferable as the Ps annihilates in the free volume holes.

The positron annihilation Doppler broadening (PADB) Spectroscopy is one of the well−known techniques for defect characterization (Dupasquier and Mills, 1995). The 511 keV annihilation line is subjected to Doppler shift which occurs as a result of the motion of the electron−positron pair. The line−shape parameters of annihilation radiation give the distribution of the longitudinal momentum component of the annihilating pair. The momentum distributions of electrons can be investigated by the DB measurement as the positrons are thermalized at the annihilation site.

The DB measurements have been performed by slow positron beams or positron-emitter radioactive sources such as ^{22}Na source. The S- (for shape), and W-parameters (for wings) (Urban-Klaehen and Quarles, 1999) are usually used for characterization of annihilation radiation. The S-parameter is defined as ratio of the net counts in the central region of the annihilation line to its total net counts. The positron annihilation with low momentum valence and unbound electrons distribution can be studied by the S−parameter due to its sensitivity to these electrons (Mackenzie et al., 1970). On the other hand, The W−parameter is more sensitive to the positron annihilation with core electrons of high momentum. This parameter is defined as the ratio between counts in the left and right wings of the peak and its total count. The equation used to calculate the S− and W−parameters are reported in Ref. (Abdel-Rahman et al., 2006b).

The information from radiochromic film dosimetry can be utilized for either qualitative or quantitative information. The film sensitivity is affected by many factor such as the photon energy, emulsion differences between film batches, experimental design and analysis tool. Several authors have investigated these variables and their effects on the relationship between the radiation absorbed dose and the optical density. An investigation of the defect structure of the active component of radiochromic films due to gamma or electrons irradiation have not been addressed in the literature at present. The main aim of this work is therefore to apply unprecedentedly PADB spectroscopy technique to investigate the changes of some structural parameters for the irradiated and unirradiated MD−55 and HD−V2 radiochromic films. In addition, correlations between the measured PADB parameters and the optical densities of the irradiated MD−55 and HD−V2 films may be possible.

2. Experimental methods

In this work, radiochromic films types MD−55 and newly released HD−V2, ISP Technologies Inc. Dosimetry Media, were used. Four and eight sets of MD−55 and HD−V2 films were prepared, respectively, with an area of 2 × 2 cm^2 for each piece. Each set contains 10 pieces. All films were marked to define their orientation and kept in a plastic bag in a light-tight envelop and stored at room temperature. Three sets of MD-55 films were given doses of 20, 40 and 80 Gy for each. Seven sets of HD-V2 films were irradiated with doses of 50, 100, 500, 800, 1000, 1500 and 2000 Gy. The MD-55 films were irradiated at room temperature and dose rate of 0.4615 Gy min^{-1} using a ^{137}Cs source. The HD-V2 films were irradiated at room temperature and dose rate of 100 Gy min^{-1} using a ^{60}Co Indian cell GC 4000 A. The irradiation process was carried out at the Gamma-Radiation Research Units, National Center for Radiation Research and Technology, Cairo, Egypt.

A 0.5 μCi ^{22}Na source was prepared using a droplet of ^{22}NaCl solution dried onto two identical Kapton foils (7.5 μm thick), which were afterward glued by epoxy glue. Four pieces of each film were placed in contact with each side of the ^{22}Na source to arrange the source and sample in a 4π configuration. The Doppler broadening line−shape parameters (S and W) were measured using a p−type HPGe detector (Ortec, GEM series) with an energy resolution (FWHM) of 1.6 keV for 1.33 MeV gamma line of ^{60}Co and relative efficiency of 25%. The amplified signals from an Ortec 570 amplifier were acquired with an Ortec 919 multichannel analyzer (MCA). The ^{133}Ba source was used for energy calibration (68 eV/channel). The DB spectra were measured in air at room temperature. About one million counts in the annihilation line were accumulated for each spectrum. The Doppler broadening spectra were analyzed using SP ver. 1.0 program (http://www.ifj.edu.pl/~mdryzek). The centroid channel with maximum counts of the 511 keV peak was carefully defined as it is a base for calculations of S− and W−parameters. The input data for this program are fixed for all spectra of the studied films. In addition, the absorbed dose profiles of MD−55 and HD−V2 films were readout using a manual radiochromic densitometer (Nuclear Associates Model 37−443) as well as a spectrophotometer (Model Labomed-UVS-2700) in the wavelength range 400−700 nm, taking the air as a baseline. All films were analyzed after 48 h from irradiation. This delay time is necessary for the films to stabilize, as recommended by several authors (AAPM , 1998; Gafchromic® MD-55).

3. Results and discussion

3.1. Optical measurements of radiochromic dosimetry films

Figs. 1 and 2 show the absorption spectra for the unirradiated and irradiated MD−55 and HD-V2 films, respectively. Both figures show that the darkening of the films increases as the γ dose increases. The active layers of the unirradiated MD−55 and HD−V2 films contain micro-crystals of a monomer. Upon exposure to ionizing radiation, the color of the active layer is changed due to the