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Performance improvement of pentacosa-diynoic acid label dosimeter for radiation processing technology

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ABSTRACT

A radiation sensitive material, 10,12-pentacosa-diynoic acid (PCDA), was incorporated into polyvinyl butyral (PVB) films to develop indicators/dosimeters for blood and food irradiation. The present study aims to improve the dosimetric performance of these previously prepared dosimeters and to extend their shelf life by the combination of a radical scavenger, propyl gallate (PG), and a UV absorber, tinuvin-p (TP). The X-ray diffraction (XRD) patterns of the dosimeters were analysed and their dosimetric characteristics were investigated by specular reflectance in the visible spectrum range of 400–700 nm. Upon irradiation, the films turn blue exhibiting two main bands around 670 and 620 nm. Their dose-response functions were fitted by a double exponential growth, 5 parameters, equation. Irradiation temperature influences the dosimeter response at 670 nm without causing thermochromic transition up to 50 °C in poly-PCDA. The useful dose range is 5–4000 Gy depending on the wavelengths of analysis and PCDA content in the films. The overall uncertainty of dose measurement is less than 6% at 2σ.

1. Introduction

Gamma radiation is applied for blood irradiation to prevent transfusion-associated graft-versus-host disease (TAGVHD) (BCSH, 1996) and for food irradiation to minimize the microbial load and to enhance the shelf life (IAEA, 2006). During irradiation treatments, dosimeters and indicators are used to follow the process and to monitor absorbed doses. For that purposes some film dosimeters have been developed and utilized, such as those based on radiation-sensitive dyes (Mai et al., 2008; Soliman and Abdel-Fattah, 2013) and conjugated diacetylene monomers (Watanabel et al., 2006; Abdel-Fattah et al., 2012).

Some conjugated diacetylene monomers are radiation-sensitive and polymerize by solid-state topochemical reaction (1,4-trans addition) upon γ-ray exposure producing deeply colored π-conjugated polydiacetylenes (Wegner, 1972; Valverde et al., 1996;). Some of these monomers were introduced as microcrystals into polymer matrixes to manufacture Gafchromic dosimetry films, which are useful for wide dose range applications (1 Gy–40 kGy) (McLaughlin et al., 1994; Devic et al., 2016; Ng et al., 2016). Gafchromic films convert into either red or blue upon γ-rays exposure (Devic, 2011). The developed colour increases proportionally with absorbed dose (Vandana et al., 2011). A new radiochromic film based on 2,4-hexadiyn-1,6-bis(*n*-butyl urethane) (HDBU) was previously prepared for high-dose range (3–150 kGy) (Abdel-Fattah et al., 2014). HDBU in the film presents a non-crystallinity structure. Thus, it randomly polymerizes upon irradiation, developing two new absorption bands at 273 nm and 285 nm. In meanwhile, when it is dispersed into polyvinyl alcohol (PVA) as a crystalline solid monomer, it polymerizes by topochemical solid-state polymerization forming a blue colored polymer at an absorbed dose of 100 Gy (Soliman et al., 2014). In this formulation, the dosimeter shows high radiation dose sensitivity and upon irradiation, it displays two main bands at 560 nm and 610 nm. Their intensity increases proportionally with absorbed dose. The expanded uncertainty of dose delivered is less than 6.5% (95% confidence level).

Pentacosa-10,12-diynoic acid (PCDA) was investigated for dosimetry purposes in gamma and UV radiation fields (Ali et al., 1996). It undergoes topochemical polymerization either by UV irradiation (Dhanabalan et al., 1996) or by γ-rays (Ali et al., 1996). In addition, PCDA was incorporated into PVB film with lower content (3–10%) for high-dose range (3–100 kGy) applications (Soliman et al., 2013). This film did not exhibit coloration upon irradiation; however, it displayed a change in the diacetylene chromofore of PCDA due to the random radiation-induced polymerization of the monomer. On the other hand, incorporating PCDA in PVB films with 20–50% produced extensive blue colour at low-dose range (15 Gy–2 kGy) (Abdel-Fattah et al., 2012). The colour change was analysed using specular reflectance colorimeter at the colour coordinates of b* colour (yellow – blue coordinate) and L* colour (light – dark coordinate). The effect of temperature on the dose-

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response function was studied; however, the effects of humidity were not discussed in the previous literature (Abdel-Fattah et al., 2012). In the present study, a tinuvin-P, a UV absorber material and a free radical scavenger (BASAF, 2016), and propyl gallate, an antioxidant for conjugated diacetylenes (Lewis et al., 2009) and a monomer stabilizer (Jackisch, 1983), were incorporated into the previously prepared films, PCDA/PVB (Abdel-Fattah et al., 2012). These additions are to improve the radiation dose sensitivity and the dosimetric performance and to extend shelf life of PCDA/PVB films. The radiation dose-responses in the present study were analysed at 480 nm, 620, and 670 nm to extend the dose range from 5 Gy to 4 kGy. In our previous investigation (Abdel-Fattah et al., 2012) the analysis at colour coordinates (b* and L* colour) did not give an ability to measure the dosimeter at extreme lower and higher dose levels; the applied dose range in this case was 15 Gy–2 kGy. The effects of environmental conditions on the dose-response and X-ray diffraction (XRD) pattern were investigated. Finally, the dosimetric results of the present dosimeter and previously prepared one (Abdel-Fattah et al., 2012) were compared.

2. Experimental and methods

The dosimeters were prepared using PVB (Pioloform BR18, average molecular weight of about 50–60 × 10^3, product of Wacker Co., USA), PCDA monomer (molecular weight 374.61, 97%, product of Fluka, USA), tinuvin p (TP) from BASF (Dispersion and Pigment division, North America) and propyl gallate (PG), product of Aldrich. Solutions containing PVB (20% wt/v) dissolved in butanol/chloroform (1/4 v/v) were used for the present dosimeters formulations. A PCDA monomer, a UV absorber of TP and a polymerization inhibitor of PG were dissolved in PVB solutions and stirred. Three formulations of the improved PCDA-PVB label dosimeter solutions were prepared with the compositions shown in Table 1. All these solutions were left tightly for 24 h under stirring to obtain well-homogenized mixtures. Then they were coated 200 µm onto self-adhesive paper (A4 size) sheets by an Automatic Film Applicator System (Braive Instrument, Belgium). The coated layers were kept at room temperature in a dark place for ~48 h for drying. Then they were cut into 2 × 2 cm^2 and stored for further analysis.

TP was used to protect the films from laboratory and fluorescent light exposure to extend the shelf life time. Meanwhile, PG was added to inhibit the dark polymerization of PCDA monomer at room temperature and thus to extend the shelf life period.

A Gamma cell 220 Excel, ^{60}Co facility (manufactured by MDS Nordion, Canada) was selected to irradiate the dosimeters at various absorbed doses in the range of 5 Gy to 4.5 kGy. A specially designed phantom of polymethylmethacrylate (PMMA) was fabricated to maintain electronic equilibrium during γ-rays exposure. This irradiation facility was supplied with a temperature control unit (National Physical Laboratory, NPL, UK) to set the temperature in the facility during irradiation and to study the effects of irradiation temperature on the dosimeter performance in the temperature range from 10 to 60 °C ± 0.1 °C. The central dose rate of the facility was calibrated by NPL using Reference Alanine Pellet Dosimeter and the dose rate at the time of this experiment was 2.2 kGy h^{-1} ± 2.6% (95% confidence level).

The optical density (-log(reflectance%)) of unirradiated and irradiated dosimeters was analysed at 400–700 nm using a specular reflectance colorimeter (Spectrocolor instrument, Dr. Bruno Lange GmbH & Co. KG Berlin, Dusseldorf, Germany). Spectrocolor is a genuine grating colorimeter measuring the visible spectral range (400 nm to 700 nm) at the interval of 10 nm. It is operating on the spectral method described in DIN 5033 using the d/8° circular viewing geometry, i.e. the sample is illuminated with polychromatic light with the optical unit observing the reflected light from a 8° position toward the sample surface.

Table 1
the compositions of the prepared label dosimeters of PVB-PCDA.

Composition ID	[PCDA] , (phr)	[TP] , (phr)	[PG] , (phr)
1 (control)	20.0	0.0	0.0
2 (improved)	20.0	7.5	5.0
3 (improved)	50.0	7.5	5.0

The X-ray diffraction (XRD) patterns of PVB films and PCDA-PVB dosimeter were recorded using Shimadzu X-ray diffractometer (XRD-6000 model, 40 kV, 30 mA) equipped with X- ray tube (Cu target). The X-ray data were recorded with continuous scanning mode and scanning speed 8 (deg/min) in the range of 5 − 45° (2θ).

To study the effect of relative humidity (RH) on the dosimeter response, the un-irradiated dosimeters were suspended in closed tubes over silica gel to adjust the dosimeters at 0% RH and over saturated salt solutions to adjust the dosimeters at different levels of RH (Greenspan, 1977); (magnesium chloride for 33%, magnesium nitrate for 53% and sodium chloride for 75%). The dosimeters were stored under these conditions for 72 h before irradiation to get equilibrium and then were irradiated simultaneously in their closed tubes at 150 Gy. After irradiation, the dosimeters were brought out at room temperature and analysed using the reflectance spectrometer.

3. Results and discussions

3.1. Effects of gamma radiation on the dosimeter response

Fig. 1 displays the spectra (400–700 nm) of unirradiated and irradiated PCDA-PVB dosimeter (composition 3) at different doses. The unirradiated dosimeters exhibit no absorption band in this range. However, under γ-irradiation, two main absorption bands develop approximately at ~676 and 620 nm characteristics of the blue colored π-conjugated poly-PCDA. The intensity of the two bands grows with increasing radiation dose until reaching saturation at higher radiation doses. The positions of these bands shift 2–4 nm toward shorter

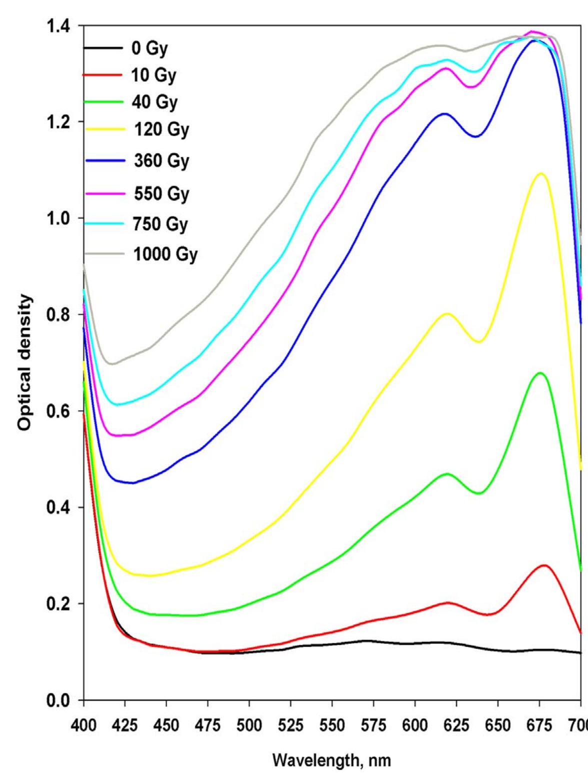

Fig. 1. Absorption spectra of unirradiated and irradiated PCDA/PVB label dosimeter (improved composition 3) at different absorbed doses.