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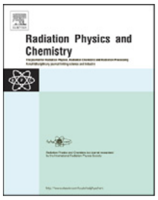

A new label dosimetry system based on pentacosa-diynoic acid monomer for low dose applications

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ABSTRACT

The dosimetric characteristics of γ-radiation sensitive labels based on polyvinyl butyral (PVB) and a conjugated diacetylene monomer, 10,12-pentacosa-diynoic acid (PCDA) have been investigated using reflectance colorimeter. Two types of labels (colourless and yellow) based on PCDA monomer were prepared using an Automatic Film Applicator System. Upon γ-ray exposure, the colourless label turns progressively blue, while the yellow colour label turns to green then to dark blue. The colour intensity of the labels is proportional to the radiation absorbed dose. The useful dose range was 15 Gy–2 kGy depending on PCDA monomer concentration. The expanded uncertainty of dose measurement of the colourless label was 6.06 (2σ).

1. Introduction

The radiation indicators may be labels, paper, inks, or packing materials, which undergo a colour change or become coloured when exposed to ionising radiation. Colour change radiation indicator labels are sometimes used on an individual item to monitor whether or not a product has been irradiated (Abdel-Rehim et al. (1985)). Label dosimeters, on the contrary, are capable of giving quantitative information regarding the dose delivered to product during routine processing (Ueno, 1988; Prussik et al., 1988; Watts and Bett, 2002).

The conjugated diacetylene monomers undergo topochemical solid state polymerisation via 1,4-addition reaction upon γ-ray exposure producing a highly conjugated coloured polymer (Wegner, 1972; Valverde et al., 1996; Hu and Li, 2002; Soliman, 2007). Such types of diacetylenes may be suitable for food irradiation dose ranges ($\approx$0.1–10 kGy) and can be used as radiation dosimeters or indicators (Patel, 1981; Lewis and Listl, 1992). Abdel-Rehim et al. (1985) have studied the dosimetric characteristics of radiation monitoring label of Allied Corporation, USA, based on some diacetylenes named 4BCMU. The radiation-induced colour change is from colourless to blue at approximate dose range 0.01–0.5 kGy. A label dosimeter system utilising a polydiacetylene as a radiochromic element printed on a barcode label and a portable hand-held computer to read the label dosimeter has been developed by Prussik et al. (1988). The reported mean standard deviation related to the measurement of the same label multiple readings and the mean readings of different label exposed to the same dose was 2.5%. The other parameters affect the uncertainty evaluation such as dose rate, and calibration relationship and environmental effect were not included in the reported mean standard deviation value by the authors.

Gafchromic films and indicators are formulated mainly from thin, active microcrystalline monomeric dispersions of diacetylenes (active layer) coated on a flexible polyester film base. The active layer turns red or blue upon γ-ray exposure producing a dye polymer. The amount of dye polymer produced, and by extension, the colour intensity increases proportionally with the increasing amount of absorbed dose in the active layer (Saylor et al., 1988; Chu et al., 1990; McLaughlin et al., 1994; Butson et al., 2001, 2003). Self-indicating, Instant, Radiation Alert Dosimeter (SIRAD) based on diacetylenes is low cost, wearable and disposal radiation dosimeter for monitoring dose ranges (0.1–1000 rads) of ionising radiation. When exposed to γ-radiation, the sensing strip SIRAD develops colour, e.g. blue or red colour instantly and the colour intensifies as the dose increases, thereby producing instantaneous information on cumulative radiation exposure in radiation personal monitoring (Patel et al., 1995; Patel et al., 2007; Cheung et al., 2007). The colouration can be used to estimate the dose with an uncertainty of 30%, especially for dosimeters exposed to low photon energy of 25.5 keV (Cheung et al., 2007). Sterin Irradiation Indicator labels (a product of International Specialty Products of Wayne New Jersey, USA) are designed to provide visual verification of irradiation treatment at 50–500 Gy dose levels using diffuse reflectance spectroscopy and have been successfully used as a quality assurance for irradiation disinfestations (Ražem, 1997). Two versions of Sterin labels (125 and 300 Gy threshold dose) were evaluated for threshold dose indication. The sensitivity

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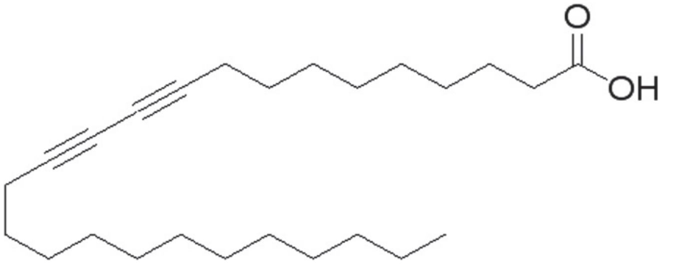
below the threshold doses was in most cases sufficient to detect ±30% variation of dose. The noise of reflectance measurements of the unirradiated label, expressed as the coefficient of variation, was about ±5%. The pre- and post-irradiation responses of this label are markedly affected by storage time. The signal of the irradiated label decays faster over the first month of storage and decreases by about 20–30% after 6 months of storage (Ražem, 1997). Such labels have been adapted to a certain extent by the blood-banking community (Ehlermann, 1997).



10, 12-pentacosadiynoic acid (PCDA)

Ali et al. (1996) have studied the irradiation of a film containing pentacosa-10, 12-diynoic acid with gamma and UV radiation for use in application of radiation processing dosimetry. This film was prepared by Langumir Blodgett (LB) technique. In the present investigation the PCDA monomer has been introduced in PVB solution for producing a radiation-sensitive layer coated on a self-adhesive paper for low dose monitoring applications. The radiation-induced colouration was analysed colorimetrically at yellow–blue coordinate (b^* colour, which is positive for yellow colours and negative for blue) and light-dark coordinate (L^* colour, which shows $L^*=100$=white and $L^*=0$=black).

The dose response functions, variation of radiation-sensitive coated layer (homogeneity of labels), and the effect of temperature on response function as well as pre- and post-irradiation stability were investigated.

2. Experimental and equipment

Two types of indicator solutions (colourless and yellow) were prepared from 20% PVB powder (Pioloform BR18, average molecular weight of about 50–60 × 10^3, product of Wacker Co., USA) mixed with PCDA monomer (molecular weight 374.61, 97%, product of Fluka, USA). The colourless PVB indicator solutions were formulated with 20 and 50 phr[a] of PCDA using solvent mixture of butanol/chloroform (1/4 v/v). The yellow PVB indicator solution was formulated with 50 phr of PCDA mixed with 0.8 phr of thymol blue (TB) indicator (molecular weight 466.59, product of CHEMAPOL, Czech Republic) sodium salt using the same solvent mixture. All solutions were well dissolved and kept at room temperature for 24 h in order to obtain a uniformly colourless (PCDA/PVB) and yellow dyed (TB/PCDA/PVB) mixed solutions. One of the purposes of the TB yellow dye is to enhance the visual contrast of the chromatic change that occurs when label is exposed to radiation. The other purpose is to shade the background of this label, a yellow colour to be easily identified when being attached to the radiation processed product boxes.

Finally, the well-mixed solutions were casted on self-adhesive paper (A4 size) sheets using an Automatic Film Applicator System (Braive Instrument, Belgium) adjusted at 200 μm. The coated layers were left for drying at room temperature at dark place for ~48 h. The coated labels were cut into 2 × 2 cm^2 and stored for dosimetric investigations.

The indicator labels were irradiated in a specially designed holder made of polystyrene using Gamma Cell 220 Excel ^{60}Co irradiation facility, GC (manufactured by MDS Nordion, Canada). The GC is equipped with a temperature control unit (manufactured and installed by National Physical Laboratory, NPL, UK) to maintain a constant temperature in the GC during irradiation in the temperature range of 10–60 ± 0.1 °C. The irradiation position was calibrated using a reference alanine dosimeter of NPL and the dose rate at the time of this experiment was 5.9 kGy h^{-1}.

The reflectance measurements of the unirradiated and irradiated labels were carried out using colorimeter (Spectro-colour instrument, Dr. Bruno Lange GmbH & Co. KG Berlin, Dusseldrof, Germany). This colorimeter 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.

3. Results and discussion

3.1. Effects of γ-ray on indicators

Upon γ-ray exposure, the colourless labels turn blue then deep blue while the yellow label turns green, then to dark blue. The radiation-induced blue colouration intensifies proportionally with increase in the amount of absorbed doses (Table 1). Due to the significant colour change upon γ-ray exposure of such labels, they can be used as radiation indicators for verifying compliance radiation processing specifications. The suitable colour change was found to start at 15 Gy depending on PCDA concentration in PVB based label (Table 1). Such labels have many advantages that are not easily scratched, easily prepared, can be manufactured with different shapes and easily to stick on cardboard product boxes. The radiation-induced blue colouration is mainly attributed to the solid state polymerisation of PCDA monomer producing a highly conjugated dye polymer, π-conjugated (Wegner, 1972; Patel, 1981; Ali et al., 1996).

3.2. Radiation response

Due to the development of blue colouration upon γ-ray exposure, the responses were investigated on two coordinates, light-dark coordinate (L^* colour) and yellow–blue coordinate (b^* colour) against absorbed doses to establish the dose–response functions. Figs. 1 and 2 show the dose response functions of colourless labels at L^* and b^*, respectively. Clearly all responses are non-linear within the useful dose range and tend to saturate at high doses. The lightness (L^* colour) decreases while the blue colour (b^* colour) increases upon γ-ray exposure. It is noted that the radiation sensitivity increases with the increasing PCDA monomer concentrations. The useful dose range was ~15 Gy–2 kGy depending on monomer

Table 1
An array of colourless and yellow labels unirradiated and irradiated to different absorbed doses.

Absorbed dose (Gy)	**Yellow label [PCDA]=50 phr**	**Colourless label [PCDA]=50 phr**	**Absorbed dose (Gy)**	**Colourless label [PCDA]=20 phr**
0	🟨	⬜	**0**	
15.1	🟩	🟦	**55.1**	🟦
105.1	🟩	🟦	**255.1**	🟦
355.1	⬛	🟦	**805.1**	🟦
755.1	⬛	🟦	**1605.1**	🟦
1005.1	⬛	🟦	**2505.1**	🟦

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[a] Phr=part per hundred parts of resin.