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Polyvinyl butyral films containing leuco-malachite green as low-dose dosimeters

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Abstract

Thin films containing leuco-malachite green (LMG) dye in polyvinyl butyral (PVB) have been developed for dose measurements of a few hundreds Gy level. The film shows significant color change in the visible range, and the sensitivity of the film to absorbed dose was enhanced by addition of chloride-containing compounds, such as chloral hydrate or 2,2,2-trichloroethanol. The film is suitable as dosimeters for dose measurements, e.g. in food irradiation and environmental protection.

Keywords: Polyvinyl butyral; Leuco-malachite green; Chloride containing compounds; ^{60}Co γ-rays; Low dose; Dosimeter; Radiation processing

1. Introduction

New thin film dosimeters are still of interest for use in radiation processing because of their convenient characteristics. The most frequently used commercially available film dosimeters are cellulose triacetate (ISO/ASTM 51650, 2005; Tamura et al., 1981), radiochromic thin film dosimeters (McLaughlin et al., 1988; ISO/ASTM 51275, 2005; Miller et al., 1988; Mai et al., 2004) and GafChromic dosimetry media (Chu et al., 1990). Most of these thin film dosimeters have been developed for use in the kGy range and only a few of them, for example, GafChromic, can be used in the lower dose range with reasonable precision.

Polyvinyl butyral (PVB) film was first studied as a dosimeter by Buenfil-burgos et al. (1983). The authors proposed a formulation of PVB and hexa (hydroxyethyl) pararose-aniline cyanide as dye but without addition of any sensitizers. This film dosimeter changes color from colorless to deep blue under irradiation at doses above several kGy. The present study aims to develop a new PVB-based film dosimeter for dose measurement of a few hundreds Gy level of γ-rays, X-rays and electron beams using leuco-malachite green (LMG) dye. Additives were tested for enhancement of radiation–coloration sensitivity, improvement of linearity of dose–response, achievement of high reproducibility and reasonable stability before and after irradiation. Chloride materials were chosen as additives because of practical easiness to control their content in films.

2. Experiment

2.1. Chemicals and preparation of film dosimeters

PVB (molecular weight $M = 90{,}000$–$120{,}000$), 2-methoxy ethanol [$CH_3OCH_2CH_2OH$], 2-ethoxy ethanol [$C_2H_5OCH_2CH_2OH$], *N*,*N*-dimethyl formamide [$HCON(CH_3)_2$], ethanol [C_2H_5OH], chloral hydrate [$CCl_3CH(OH)_2$], 2,2,2-trichloroethanol [CCl_3CH_2OH] and LMG [$C_6H_5CH[C_6H_4N(CH_3)_2]_2$], were all supplied by Sigma-Aldrich Corporation, of analytical grade and were used without any further purification.

The PVB powder (grain size is about 0.3 mm) was dissolved in mixed solvent of 2-ethoxy ethanol and 2-methoxy ethanol in a volume ratio of 50:50 to prepare

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a stock gel polymer solution with solid content of PVB to be 0.08 g/ml. The amounts of each chemical in the formulation of the PVB–LMG film are listed below:

PVB	10 g
2-Methoxy ethanol	50 ml
2-Ethoxy ethanol	50 ml
N,*N*-Dimethylformamide	10 ml
1-Butanol	10 ml
Leuco-malachite green (LMG)	0.3–0.7 g (3–7 wt% in solid content of PVB)
Chloral hydrate/2,2,2-trichloroethanol	1–5 g (10–50 wt% in solid content of PVB)

The stock polymer solution was well stirred at a temperature of 45 °C for 24 h to completely dissolve the PVB powder. LMG was thoroughly dissolved in 10 ml of *N*,*N*-dimethyl formamide before adding it to the PVB gel solution and stirred for 10 h at room temperature (25 ± 5 °C). Chloral hydrate or 2,2,2-trichloroethanol was then added to the solution in different amounts and also well stirred for at least 10 h at room temperature to get a homogeneous solution. The final gel polymer solution has a viscosity of 1850 ± 50 mN s/m^2 at temperature of 25 °C that is suitable for preparing thin films by casting or coating process. In the casting process, the PVB thin films were made by pouring the dyed solution on a perfectly leveled glass plate and dried for 72 h at room temperature. The relative humidity was 65 ± 15% during preparation. The thickness of the film was controlled to be in the range 50–70 μm by weighing the amount of solution and limiting the casting area on the glass plate. The films were stripped carefully from the glass plate and were kept for drying in an incubator at 45 °C in order to remove residues of the solvents completely. Finally, the films were cut into pieces of 1 cm width and 3.5 cm length so as to be suitable to the sample holders of common UV–vis spectrophotometers. Each piece of PVB–LMG film was sealed into a small dark plastic pouch and stored under laboratory condition.

2.2. Irradiation and readout equipment

The PVB–LMG films were irradiated to different doses using a ^{60}Co γ-ray source having radioactivity of 4 TBq. The absorbed dose rate at the irradiation position was measured to be 150 Gy/h using alanine dosimeter (Hitachi Cable Co. Ltd., ISO/ASTM 51607). At each dose level, three films were sandwiched together between two polymethyl-methacrylate (PMMA) plates with 3 mm thickness in order to establish secondary charged particle equilibrium.

A 400 keV electron accelerator (Russian Academy of Science, URT-0.4, pulse 50 ns, 1 Hz) (Kotov and Sokovnin, 1997) was used for irradiation and to demonstrate the dose distribution measurement.

The optical spectra (400–800 nm) and the absorbance of the irradiated films at 629 nm were measured by a UV–vis spectrophotometer (HITACHI U-3310) with a bandwidth of 2 nm. The uncertainty in wavelength setting was ±0.01 nm, and the fluctuation in absorbance measurement was ±0.001 for an absorption range of 0–1.0. The thickness of the films was measured after optical absorbance measurement using a digital gauge with a precision of ±1 μm.

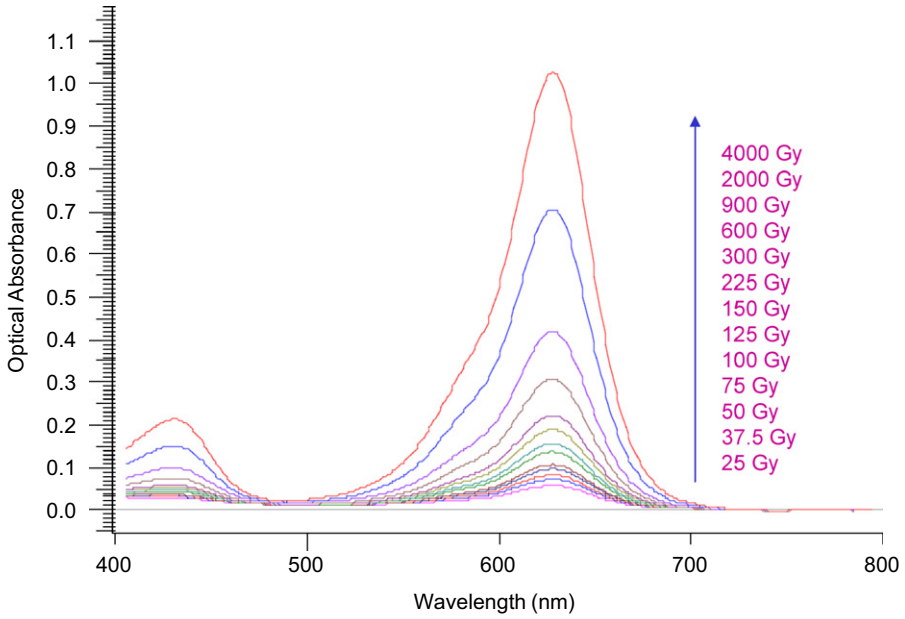

Fig. 1. Optical absorption spectra of PVB–LMG films irradiated by γ-rays to different doses.