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Radiation-induced color bleaching of methyl red in polyvinyl butyral film dosimeter

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ABSTRACT

Radio-chromic film based on polyvinyl butyral (PVB) containing different concentrations of methyl red (MR) dye for 0.125, 0.25 and 0.5 mM has been introduced as high dose dosimeter. The dosimeters were irradiated with gamma ray from ^{60}Co source at doses from 5 to 150 kGy. UV/vis spectrophotometry was used to investigate the optical density of unirradiated and irradiated films in terms of absorbance at 497 nm. The dose sensitivity of MR-PVB film dosimeter increases strongly with increase of absorbed dose as well as increase of concentrations of MR dye. The effects of irradiation temperature, relative humidity, dose rate and the stability of the response of the films after irradiation were investigated and found that these films could be used as routine dosimeter in industrial radiation processing. The useful dose range of developed MR-PVB film dosimeters is in the range of 5–100 kGy.

1. Introduction

Radio-chromic film dosimeters based on the radiation-induced color bleaching of organic dyes have been extensively investigated for high dose dosimetry in radiation processing (McLaughlin, 1970; Miller, 1986; Kovacs et al., 1998; Barakat et al., 2001; Chen et al., 2008). Ajji (2006) investigated gamma radiation-induced decoloration of methyl red in alkaline and acidic aqueous solutions. The response in alkaline medium decreased exponentially with increase of absorbed dose in the range between 50 and 6000 Gy, while the response of the acidic dye solution dosimeter decreased linearly with increase of absorbed dose up to 200 Gy.

Radiation-induced color bleaching of polyvinyl alcohol (PVA) film containing methyl red dye was investigated in the dose range from 10 to 55 kGy (Bhat et al., 2007). When the film was subjected to gamma radiation, the color of MR-PVA film changed from yellow to colorless and its complete decoloration occurred at irradiation dose of 30 kGy. It was reported that these films can be used as high dose routine dosimeter. Polyvinyl butyral (PVB) dosimeter with different dye in the dose range from 1 to 4 kGy was developed (Abdel-Fattah and El-Kelany, 1998). This film was based on PVB binder containing acidic sensitive dye (bromophenol blue) and chloral hydrate. The response of the films increases significantly with increased dose in the mentioned range and showed good stability after irradiation as well as a negligible relative humidity effect in the range from 10 to 70%.

In this work, a composition of methyl red polyvinyl butyral (MR-PVB) is introduced as a radio-chromic film dosimeter. Since radio-chromic films used in routine dosimetry systems are usually influenced by the environmental conditions such as relative humidity, irradiation temperature, dose rate and post-irradiation time (McLaughlin et al., 1989; Farah et al., 2004), therefore this study investigated the effect of these parameters on the performance of MR-PVB film dosimeters in the dose range from 5 to 150 kGy.

2. Experimental

Stock solutions containing 0.125, 0.25 and 0.5 mM MR dye were prepared by dissolving different weights of methyl red, $C_{15}H_{15}N_3O_2$ (Alfa Aesar, USA) in 100 ml 96% ethanol in a 150 ml volumetric flask. The solutions were stirred in the dark for 24 h at room temperature in order to obtain homogeneous dye stock solutions. Polyvinyl butyral (PVB) solutions were prepared by dissolving 16 g of PVB powder (MW = 36,000 g/M, Wacker Chemicals, USA) in 160 ml ethanol at 60 °C. The solution was magnetically stirred at this temperature for 4 h and then followed by stirring for 24 h at room temperature. The PVB solutions were then divided into 40 ml samples and different concentrations of MR solutions (i.e. 0.125, 0.25 and 0.5 mM) were added to 40 ml PVB solution. Mixtures were stirred continuously in the dark for 24 h at room temperature using

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a magnetic stirrer in order to obtain a uniformly dyed PVB solution. MR-PVB solutions were poured onto a horizontally levelled polystyrene plate and dried at room temperature for about 72 h. Films were peeled off and cut into $1 \times 3\ cm^2$ pieces, dried, stored and prepared for irradiation. The drying is completed when the weight of the films was constant. The mean thickness of the obtained films is 0.125 ± 0.011 mm, which was measured by high precision digital micrometer (Mitutoyo-Japan) and stored in special envelopes to protect the films from sunlight, fluorescent light, moisture and dust.

Dyed PVB films were irradiated with ^{60}Co gamma radiation (Gammacell-220 irradiator supplied by MDS Nordion, Canada) at dose rate of 11.98 kGy/h. The temperature during irradiation was set with an air chiller system (Turbo-Jet, Kinetics, USA). The dose rate of the gamma source was measured by using ferrous sulfate (Fricke) dosimeter (ASTM Standard Practice E1026, 2004). The irradiations were conducted at various temperatures. At each dose point, three films were sandwiched together between two polystyrene (PS) blocks with 6 mm thickness in order to establish secondary charged particle equilibrium and the average is reported. The PS blocks were positioned at the center where absorbed dose is uniform. A range of 12–75% relative humidity levels was used to study the effect of humidity on the performance of MR-PVB film dosimeters during irradiation. These humidity levels were achieved using the following saturated salt solutions: LiCl (12%), $MgCl_2 \cdot 6H_2O$ (34%), $Mg(NO_3)_2 \cdot 6H_2O$ (55%) and NaCl (75%) according to the technique devised by Levine et al. (1979). The films irradiated in a given humidity environment were kept in the same environment for 3 days before irradiation to ensure equilibrium conditions. Electron beam irradiation was conducted at Sure Beam Middle East Corp. (SME) irradiation facility in Riyadh, Saudi Arabia with two electron beam accelerators positioned vertically (tower and bit) at dose rate of 1 kGy/s and at room temperature. The electron beam parameters are listed in Table 1. Alanine pellet dosimeters were used as a reference dosimeter irradiated along developed films in accordance with ISO/ASTM 51607 (2004). Alanine pellet dosimeters are traceable to National Institute of Standard and Technology (NIST) in USA. Alanine dosimeters are measured by E-Scan EPR spectrometer (Bruker Biospin Company, Germany). Similar to gamma irradiation, three films were sandwiched together between two PS blocks in order to maintain electronic equilibrium during EB irradiation. The PS blocks were placed on the tray of conveyor system of EB accelerator. The absorption spectra of the irradiated MR-PVB films were measured with an UV/VIS spectrophotometer (model Lambda 20, from Perkin-Elmer, USA)in the wavelength range of 350–650 nm.

In general, evaluation of MR-PVB followed ASTM standard guide for performance characterization of dosimeters and dosimetry systems for use in radiation processing, ASTM E2701-09 (ASTM Standard Guide E2701, 2009).

Table 1
Electron beam system operating parameters.

Parameter	Tower	Pit
Beam energy (MeV)	10	10
Average beam current (mA)	1.43	1.6
Average beam power (kW)	14.3	16
Scan magnet current (A)	233	200
Scan width (cm)	120	120
Pulse per scan	64	64
Scan frequency (Hz)	5.05	5.26
Pulse repetition rate (Hz)	288	302

3. Results and discussion

3.1. Effect of MR dye concentration on the performance of the film

The effect of the dye concentrations on the response of the MR-PVB films was investigated by preparing different compositions of MR-PVB films. The dose response curves were established in terms of change in absorption peak measured at 497 nm per thickness in mm (specific value) versus the absorbed dose. The absorption spectra of unirradiated as well as irradiated MR-PVB films were measured in the range 350–650 nm. Lower concentrations of MR were used initially and found not useful. Fig. 1 shows the absorbance spectra of 0.5 mM of MR in the dose range 0–150 kGy. Dose response of MR-PVB films is shown in Fig. 2. The color bleaching of MR-PVB film increases gradually with increasing absorbed dose up to 100 kGy, then it tends to saturate up to 150 kGy, which can be seen from decrease of the individual relative absorbance–dose curve. As the absorbed dose increases, more hydrated electrons and free radicals are generated leading to breakage of azo group of MR dye, resulting in the disappearance of chromophore (Bhat et al., 2007). The results show that dose sensitivity as well as the dose range increases with increase of dye concentration, indicating that films containing higher concentrations of the MR dye are more suitable for higher dose

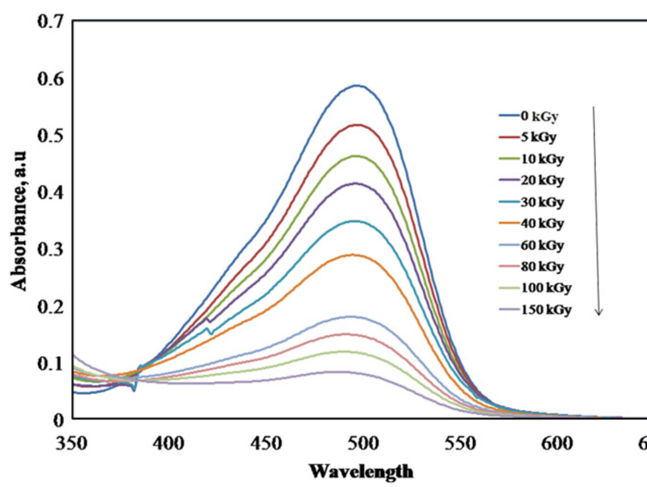

Fig. 1. The absorption spectra of unirradiated as well as irradiated of 0.5 mM MR-PVB films.

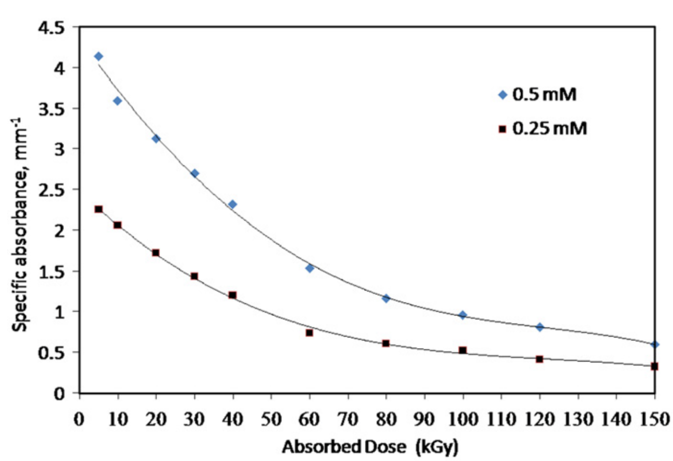

Fig. 2. Specific absorbance of MR-PVB film dosimeters containing 0.25 and 0.5 mM MR as a function of absorbed dose.