Radiation Measurements 47 (2012) 1005-1008

Contents lists available at SciVerse ScienceDirect



Radiation Measurements

journal homepage: www.elsevier.com/locate/radmeas

Improved performance of nitro-blue tetrazolium polyvinyl butyral high dose film dosimeters

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HIGHLIGHTS

- ▶ This manuscript relates to radio-chromic dosimeter for use in high dose radiation.
- ► Radiolytic reduction of nitro blue tetrazolium (NBT) results in a water insoluble formazan.
- ▶ NBT-PVB films were prepared in 2-methoxyethanol solution with variable concentrations of NBT dye.
- ► The dose sensitivity of NBT-PVB films increases strongly with increase of NBT dye concentration.
- ▶ NBT-PVB films slightly depends on relative humidity and irradiation temperature.

ARTICLE INFO

Article history: Received 6 March 2012 Received in revised form 26 June 2012 Accepted 20 July 2012

Keywords: Radio-chromic film Nitro blue tetrazolium Polyvinyl butyral 2-Methoxyethanol Humidity Irradiation temperature

ABSTRACT

Improved performance in terms of dependence on humidity and irradiation temperature for radiochromic film dosimeters with high dose range was achieved based on radiation-induced reduction of NBT2⁺. NBT-PVB film dosimeters containing different concentrations of NBT dye from 8 to 12 mM were prepared in a solution of 2-methoxyethanol. The dosimeters were irradiated with γ -ray from ⁶⁰Co source at doses up to 100 kGy. UV/VIS spectrophotometer was used to investigate the optical density of unirradiated and irradiated films in terms of absorbance at 525 nm. The absorbance increases with absorbed dose up to 100 kGy for NBT-PVB film dosimeters. The dose sensitivity of NBT-PVB film increases strongly with increase of concentrations of NBT dye. The effects of irradiation temperature, humidity, and the stability of the response of the films after irradiation were investigated. The stability of film dosimeters after irradiation was very high up to 60 days.

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1. Introduction

Quality control of radiation processing depends strongly on availability of reliable film dosimeters, therefore many types of films are used for radiation dosimetry such as Radio-chromic films of various types (Al Zahrany et al., 2011; Rabaeh et al., in press). Tetrazolium salts were recently found to be useful for chromatic dosimetry purposes. Tetrazolium salts are heterocyclic organic compounds, which upon irradiation yield highly colored water insoluble formazans (Al-Sheikhly et al., 1998). Ionizing radiation induced reduction of tetrazolium salts in polyvinyl alcohol (PVA) film dosimeters such as 2,3,5-Triphenyltetrazolium chloride TTC-PVA film dosimeter (Kovács et al., 1995), tetrazolium violet TV- PVA film dosimeter (Emi-Reynolds et al., 2007), nitro blue tetrazolium chloride NBT-PVA film dosimeter (Kovács et al., 1999; Moussa et al., 2003; Rabaeh et al., 2012) were investigated. It was found that NBT can be used as a radiation dosimeter in the range between 5 and 50 kGy, but the major disadvantages of previous studies was the strong effect of high humidity environment (amount of water content during irradiation) on the performance of these dosimeters. Due to the different environmental conditions between calibration and practical use, this disadvantage leads to limitation of using these films as a routine dosimeter in gamma radiation processing.

Basfar et al. (2011) was the first group who reduced the high effect of humidity on this type of film by introducing nitro blue tetrazolium chloride polyvinyl butyral (NBT-PVB) film dosimeter prepared in ethanol. PVB is a resin usually used for applications that require strong binding, optical clarity, and flexibility. PVB does not dissolve in water, so it is less affected by humidity. The low

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homogeneity of NBT-PVB solution especially at concentration more than 5 mM of NBT dye leads to limited performance of NBT-PVB films prepared in ethanol. The concentration of NBT dye of this film was limited to 5 mM with an effective dose response at relatively large thicknesses (more than 200 μ m).

2-methoxyethanol is introduced in the current work to increase the solubility of both NBT and PVB and produce a high concentration NBT-PVB film dosimeter with an effective dose response at lower thickness (100 μ m). Therefore, the influence of humidity and irradiation temperature was reduced further. The dose range of NBT-PVB film dosimeters is also increased up to 100 kGy.

2. Experimental

Polyvinyl butyral (PVB) solutions were prepared by dissolving 9 g PVB powder (Mw = 36,000 g/M, Wacker, USA) in 110 ml 2methoxyethanol at 45 °C. The solution was magnetically stirred at this temperature for 4 h and then left to cool down to room temperature. After cooling down to room temperature, PVB solution was divided into three volumes. PVB composites were prepared by dissolving different concentrations of NBT (i.e. 8, 10, and 12 mM) in the three volumes of PVB solutions, respectively. Mixtures were stirred continuously for 24 h using a magnetic stirrer in order to obtain uniformly dyed PVB solutions. NBT-PVB solutions were poured onto a high leveled horizontal glass plates and dried at room temperature for about 72 h. Films were peeled off and cut into 1×3 cm pieces, dried, stored and prepared for irradiation. The drying is completed when the weight of the films is constant. The films were protected from sunlight, fluorescent light. moisture and dust by storing them in a paper envelope and wrapping them with a black plastic tape. The thickness of the film is $100 \pm 5 \ \mu m$ with very good dye homogeneity.

NBT-PVB films were irradiated with ⁶⁰Co 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 using a 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) plates with 6 mm thickness in order to establish secondary charged particle equilibrium and the average is reported. No significant differences in their characteristics were found during measurements. Relative humidity levels in the range of 12–75% were used to study the effect of humidity on the performance of NBT-PVB film dosimeters during irradiation. These humidity levels were achieved using the following saturated salt solutions: LiCl (12%), MgCl₂.6H₂O (34%), Mg(NO₃)₂.6H₂O (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.

The absorption spectra of the irradiated NBT-PVB films were measured with a UV/VIS spectrophotometer (model Lambda 20, from Perkin–Elmer, USA) in the wavelength range of 350–650 nm. In general, evaluation of NBT-PVB followed ASTM standard guide for performance characterization of dosimeters and dosimetry systems for use in radiation processing, ASTM E2701-09 (ASTM Standard Guide E2701-09).

3. Results and discussion

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

The effect of the dye concentrations on the response of the film dosimeters was investigated in different compositions of NBT-PVB

films. A set of three films was used for each irradiation dose. Fig. 1 shows the absorbance spectra of 12 mM of NBT-PVB film dosimeter in the dose range 0–100 kGy. The absorption maximum is located at about 520 nm with a shoulder at about 575 nm due to the absorbance of mono-formazan (Kovács et al., 1999). It was observed that the absorbance on the long wavelength side of the spectrum (about 575 nm) gets stronger with increasing absorbed dose, as a result of formation of di-fomazan species from reduction of mono-formazan species. The dose response curves were established in terms of change in absorption peak measured at 525 nm per thickness in mm ($\Delta A = A_x - A_0$) versus the absorbed dose, where A_x and A_0 are absorbance values at 525 nm for irradiated and un-irradiated films (Ebraheem et al., 2007). Dose response of PVB films are shown in Fig. 2(a and b), respectively.

Fig. 2 shows the specific absorbance values of 8, 10 and 12 mM NBT-PVB film dosimeters normalized with respect to value of 100 kGy. The dose response of NBT-PVB film increases with increase of dose, which can be seen from an increase of the individual relative absorbance-dose curves (see Fig. 2). As the dose increases, more hydrated electrons and free radicals are generated leading to breakage of $N-N^+$ bonds, resulting in an increase in the formation of colored formazan. The results show that dose response increases with increase of dye concentration, indicating that NBT-PVB film dosimeters containing higher concentrations of the NBT dye are more suitable for high dose dosimetry. This is in a good agreement with previous studies for different types of tetrazolium salt dosimeters (Bielski et al., 1980; Moussa et al., 2003 and Basfar et al., 2011). The dose sensitivity was taken from the slope of the linear curve of dose D versus specific absorbance in Fig. 2(b). The results show that the maximum difference of sensitivity reached 95% for different dye concentrations from 8 to 12 mM NBT.

3.2. Effect of irradiation temperature on performance of NBT-PVB films

The effect of irradiation temperature on the response of NBT-PVB films was investigated by irradiating film samples containing 10 mM NBT dye to 5, 10 and 20 kGy in the temperature range of 10–40 °C. A set of three films was used for each temperature. The results show that films depend on irradiation temperature where the maximum difference of specific absorbance reaches 16% for different irradiation temperatures from 10 to 40 °C at 20 kGy. Accordingly, the response of the films has to be corrected under



Fig. 1. Absorbance of 12 mM of NBT-PVB film dosimeters as a function of wavelength for various absorbed doses.

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