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Dosimetry characterization of nitro-blue tetrazolium polyvinyl butyral films for radiation processing

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ABSTRACT

Nitro-blue tetrazolium polyvinyl butyral film dosimeters (NBT-PVB) were prepared and investigated based on radiation-induced reduction of NBT^{2+}. NBT-PVB film dosimeters containing different concentrations of NBT dye from 1 to 5 mM were prepared in a solution of ethanol. The dosimeters were irradiated with γ-ray from ^{60}Co source at doses from 5 up to 55 kGy. UV/vis spectrophotometry was used to investigate the optical density of unirradiated and irradiated films in terms of absorbance at 529 nm. The absorbance increases with absorbed dose up to 55 kGy for NBT-PVB film dosimeters. The dose sensitivity of NBT-PVB film increases strongly with an increase in concentrations of NBT dye. The effects of irradiation temperature, humidity, dose rate and the stability of the response of the films after irradiation were investigated. The influence of irradiation temperature and humidity on the performance of the film was reduced significantly due to the use of PVB as a binder containing NBT dye.

1. Introduction

Several techniques were developed to measure absorbed dose by using appropriate dosimeter systems. Accordingly, scientists are continuing the search for new dosimeters, which can meet demands of new radiation processing applications. An ideal dosimeter would be simple to use and physically sturdy suitable for medical and industrial use on a routine basis. The dosimeter must also have an acceptable accuracy and precision with a relative ease of handling and data analysis (Kantz and Humpherys, 1977).

Radio-chromic film dosimeters depend on permanent change in color due to radiation-induced chemical changes. This color change is proportional to absorbed dose. Nowadays, there are various film types with various chemicals, where the competition between them depends on their accuracy, the limits of lower and higher doses to be measured and their behavior under different environmental conditions such as temperature and humidity.

A dosimeter similar to film of present work based on polyvinyl butyral (PVB) but with different dye and low dose range (up to 4 kGy) was developed (Mai et al., 2008). This film is based on PVB binder containing leuco-malachite green dye as a low dose dosimeter. The film showed linear response for radiation exposure in the mentioned range and showed good stability after irradiation and is considered as a promising dosimetry system. As a result of limited dose range (up to 4 kGy), this film is not suitable for high dose application such as sterilization and food irradiation.

Tetrazolium salts were recently found to be useful for dosimetry purposes. These salts are heterocyclic organic compounds, which on irradiation yield highly colored water insoluble formazans due to radiolytic reduction (Auclair and Voisin, 1985). Different tetrazolium salts such as 2,3,5-triphenyltetrazolium chloride (TTC) (Kovács et al., 1996) , tetrazolium blue (TB) (Al-Sheikhly et al., 1998), nitro-blue tetrazolium (NBT) (Kovács et al., 1999; Moussa et al., 2003) and tetrazolium violet (TV) (Emi-Reynolds et al., 2007a, 2007b) were investigated using polyvinyl alcohol (PVA) as a binder. It was found that films containing tetrazolium salts can be used as radiation dosimeter in the range of 5–50 kGy but the major disadvantage of previous studies is the strong effect of high humidity environment on the performance of these dosimeters. Therefore, nitro-blue tetrazolium polyvinyl butyral (NBT-PVB) film dosimeter is introduced in this study. This work also aims to investigate the effects of humidity, irradiation temperature and dose rate on the performance of NBT-PVB film for dose measurements as routine dosimeters in the dose range of 5–55 kGy.

2. Experimental

Stock solutions containing 1, 2.5 and 5 mM NBT dye were prepared by dissolving different weights of nitro-blue tetrazolium chloride ($NBTCl_2$; product of Biosynth, USA) in 100 ml of 96% ethanol in a 150 ml volumetric flask (Table 1).

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Table 1
Various concentrations of NBT dye solution.

Stock solution no.	Concentration (mM)
1	5
2	2.5
3	1

The solutions were stirred at room temperature for 3 h to ensure homogeneous dye stock solutions. Polyvinyl butyral (PVB) solutions were prepared by dissolving 18 g of PVB powder (Mw=36,000 g/M, Wacker Chemicals, USA) in 180 ml ethanol at 60 °C. The solution was magnetically stirred at this temperature for 4 h and then left to cool down to room temperature. The PVB solutions were then divided into 60 ml samples and different concentrations of NBT solutions (i.e. 1, 2.5 and 5 mM) were added to 60 ml PVB solution. Mixtures were stirred continuously for 24 h using a magnetic stirrer in order to obtain a uniformly dyed PVB solution. NBT-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 $1\times3\,cm^2$ pieces, dried, stored and prepared for irradiation. The drying is completed when the weight of the films was constant. The films were protected from sunlight, fluorescent light, moisture and dust by storing them in small paper envelop and wrapping them with black plastic tape.

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 using a ferrous sulphate (Fricke) dosimeter (ASTM Standard Practice E1026, 2004). The irradiations were conducted at various temperatures. At each dose point, three films were irradiated and measured and the average is reported.

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%), $MgCl_2\cdot6H_2O$ (34%), $Mg(NO_3)_2\cdot6H_2O$ (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.

The absorption spectra of the irradiated NBT-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 NBT-PVB followed ASTM standard guide for performance characterization of dosimeters and dosimetry systems for use in radiation processing, ASTME2701-09 (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 concentration on the response of the NBT-PVB films was investigated by preparing different compositions of NBT-PVB films. The dose response curves were established in terms of change in specific absorbance (i.e. absorbance measured at the absorption peak, 529 nm, per unit thickness) ΔA ($\Delta A=A_x-A_0$) versus the absorbed dose, where A_x and A_0 are the absorbance values at 529 nm for irradiated and unirradiated films. The dose response curves of the NBT-PVB films are shown in Fig. 1.

For all concentrations of NBT dye, the dose response of the NBT-PVB film increases with the increase in dose (see Fig. 1). As the dose increases, more hydrated electrons and free radicals are generated leading to the breakage of $N-N^+$ bonds of NBT, resulting in an increase in the formation of formazan. The results show that the dose response increases with the increase in dye concentration, indicating that films containing higher concentrations of the NBT dye are more suitable for high dose dosimetry. This observation is in good agreement with previous studies for different types of tetrazolium salt dosimeters (Bielski et al. 1980; Moussa et al. 2003; Emi-Reynolds et al. 2007a).

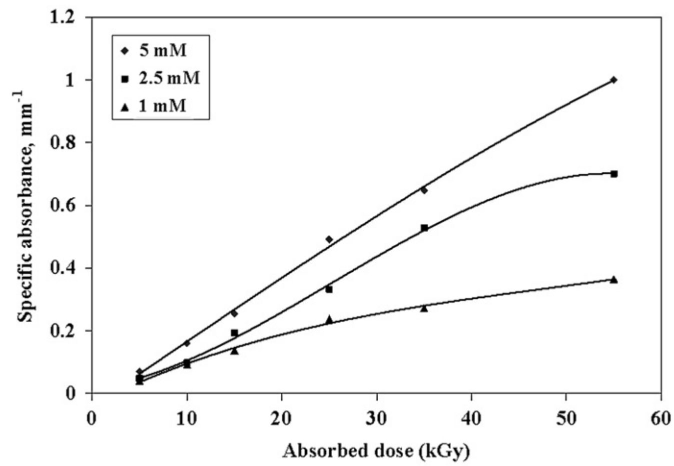

Fig. 1. Specific absorbance at 529 nm of 1, 2.5 and 5 mM NBT-PVB film dosimeter as a function of absorbed dose.

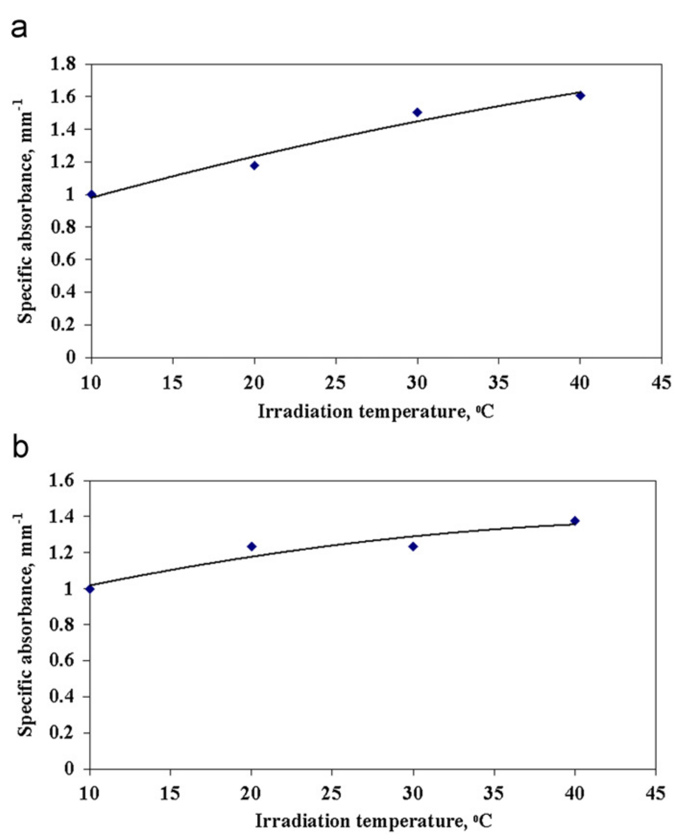

Fig. 2. Specific absorbance of 5 mM NBT-PVB film dosimeters normalized to the value measured at 10 °C as a function of irradiation temperature for (a) 5 kGy and (b) 10 kGy.

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

The effect of irradiation temperature on the response of NBT-PVB films was investigated by irradiating film samples containing 5 mM NBT dye to 5 and 10 kGy in the temperature range of 10–40 °C. The variation in absorbance of the films was normalized with respect to the measured value at 10 °C (see Fig. 2a and b). The results show that the response of the films depends on the irradiation temperature; therefore in-plant calibration or verification exercises are suggested (Sharpe and Miller 1999). This observation is in agreement with previous work by Farah et al. (2004).

3.3. Effect of humidity on the performance of NBT-PVB films

The effect of humidity on the NBT-PVB film dosimeters was investigated by storing film samples containing 5 mM NBT dye in vials in different humidity levels (12%, 34%, 55% and 74% relative humidity) for three days to establish equilibrium in accordance with ASTM E2701-09 (ASTM Standard Guide E2701-09).

The films were then irradiated in the same vials to10 and 20 kGy. The variation in absorbance of the irradiated films (10 and 20 kGy) was normalized with respect to the value measured at 12% relative humidity (see Fig. 3a and b). The results show that the dose response increases reasonably with an increase in relative humidity – especially over 50% – compared to previous studies of NBT-PVA film dosimeters (Moussa et al., 2003).

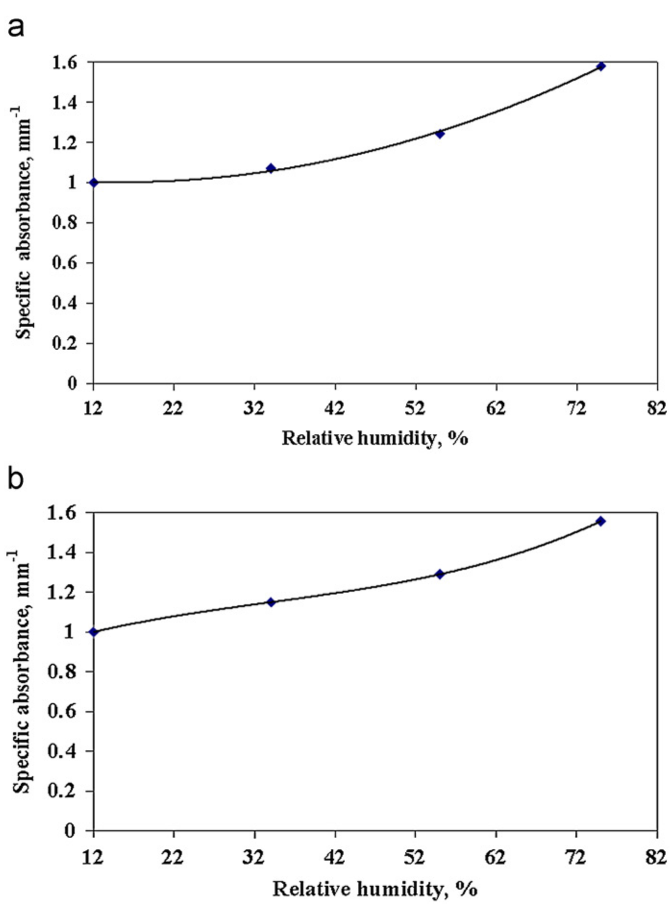

Fig. 3. Specific absorbance of 5 mM NBT-PVB film dosimeters normalized to the value measured at 12% relative humidity at (a) 10 kGy and (b) 20 kGy.

3.4. Stability of NBT-PVB film dosimeter after irradiation

Similar to the other types of radio-chromic dosimeter films, the stability of NBT-PVB films was tested by measuring the absorbance of NBT film containing 5 mM NBT. The films were irradiated to 5 and 10 kGy and kept under normal laboratory conditions of

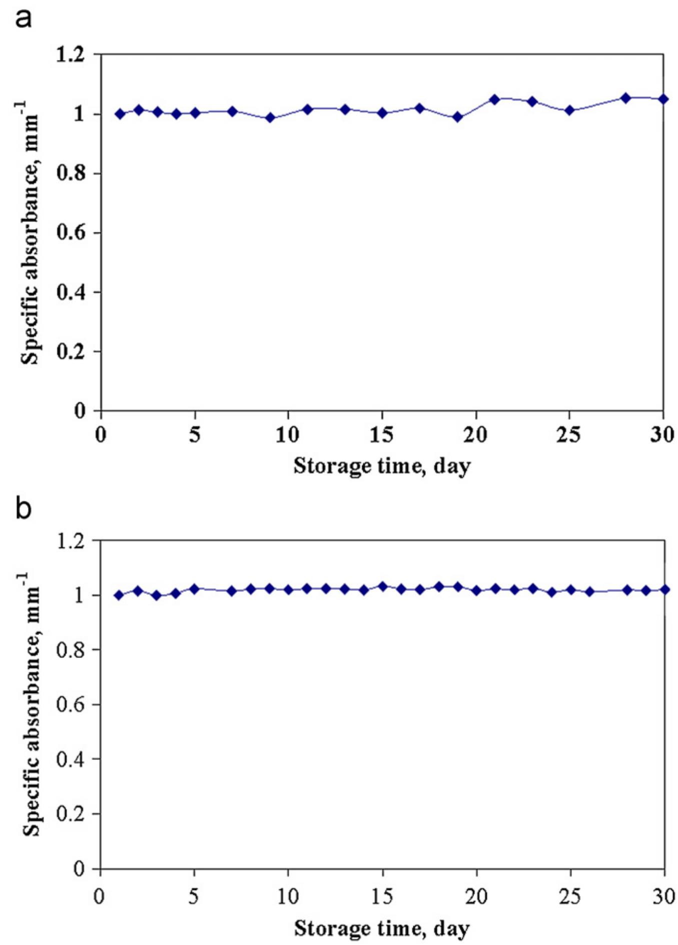

Fig. 4. Specific absorbance of 5 mM NBT-PVB film dosimeters normalized to the value measured at one day after irradiation for different absorbed doses: (a) 5 kGy and (b) 10 kGy.

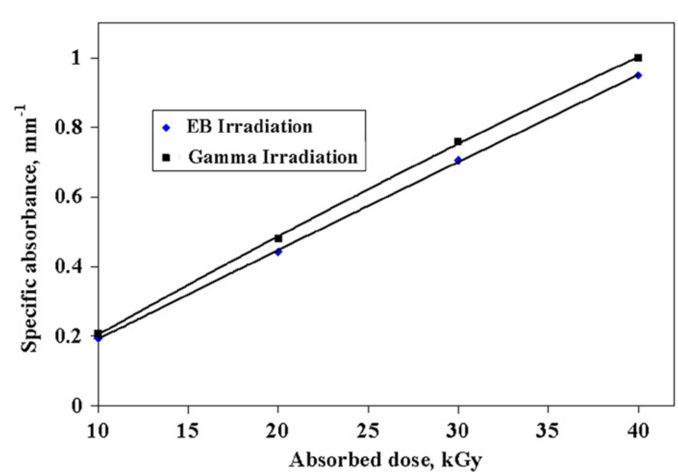

Fig. 5. Specific absorbance of 5 mM NBT-PVB film dosimeters irradiated with gamma and electron radiation to different absorbed doses.

Table 2
Absorbance of NBT-PVB film with precision values.

Film no.	Absorbance (au)		
	5 kGy	15 kGy	35 kGy
1	0.0625	0.223	0.648
2	0.0598	0.23	0.623
3	0.0642	0.213	0.656
Average	0.062167	0.222	0.642333
SD	0.002219	0.008544	0.017214
Precision (%)	±3.6	±3.8	±2.7

temperature and humidity in dark in accordance with ASTM E2701-09 (ASTM Standard Guide E2701-09). The optical density of the irradiated NBT-PVB films was measured every 24 h using UV/vis spectrophotometer for 30 days after irradiation. The results show no change (less than $\pm 3\%$; 1σ) in the specific absorbance of the film up to 30 days (see Fig. 4a and b) . This result is in agreement with previous work by Kovács et al. (1995).

3.4.1. *Effect of dose rate*

The effect of dose rate on the response of NBT-PVB film dosimeters was investigated by using a dose rate of 11.98 kGy/h from a ^{60}Co source and an electron beam accelerator with a dose rate of 15 kGy/s. The film dosimeters were irradiated at a relative humidity of 50% and at a temperature of 25 °C for absorbed doses of 10, 20, 30 and 40 kGy. It was found that there is no significant effect of dose rate on the response of the NBT-PVB film dosimeters (see Fig. 5). This result is in agreement with previous work by Moussa et al. (2003).

3.4.2. *Reproducibility*

Three sets of 5 mM NBT-PVB film dosimeters were prepared separately and irradiated to different doses as presented in Table 2. The results demonstrated that the absorbance has good precision values with standard deviation values ($\sigma \leq 4\%$), indicating that the film preparation has a good degree of reproducibility for radiation processing industry.

4. Conclusions

NBT-PVB film dosimeters were prepared with variable concentrations of NBT dye from 1 to 5 mM. Based on the systematic evaluation of the dosimetric properties of these films performed, it was found that the film is useful for routine dosimetry in industrial radiation processing. The absorbance increases with absorbed dose in the dose range of 5–55 kGy for NBT-PVB films prepared in ethanol solution. The dose sensitivity increases strongly with the increase in the NBT dye concentration. The response of NBT-PVA film increased significantly with increase in relative humidity and irradiation temperature (Moussa et al., 2003), while the sensitivity of the NBT-PVB film increased reasonably with increase in relative humidity and irradiation temperature, which enhanced directly the stability of these films. The response of the films has to be calibrated or verified under actual processing conditions. Suitable stability of the response of the film dosimeters after irradiation was found up to 30 days. The effect of dose rate on the response of NBT-PVB film dosimeters was investigated and found that there is no appreciable effect of dose rate on NBT film dosimeters. The reproducibility of NBT-PVB film dosimeters was within $\pm 4\%$.

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