Accepted Manuscript

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PII: S2211-3797(17)32408-7
DOI: https://doi.org/10.1016/j.rinp.2018.01.050
Reference: RINP 1207

To appear in: Results in Physics

Received Date: 3 December 2017
Revised Date: 4 January 2018
Accepted Date: 21 January 2018

Please cite this article as: Aldweri, F.M., Abuzayed, M.H., Al-Ajaleen, M.S., Rabaeh, K.A., Characterization of Thymol Blue Radiochromic Dosimeters for High Dose Applications, Results in Physics (2018), doi: https://doi.org/10.1016/j.rinp.2018.01.050

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Characterization of Thymol Blue Radiochromic Dosimeters for High Dose Applications

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Abstract
Thymol blue (TB) solutions and Thymol blue Polyvinyl Alcohol (TB-PVA) films have been introduced as Radiochromic dosimeter for high dose applications. The dosimeters were irradiated with gamma ray (60Co source) from 5 to 30 kGy for film, and from 0.150 kGy to 4 kGy for solution. The optical density of unirradiated and irradiated TB solution as well as TB-PVA film dosimeters were studied in terms of absorbance at 434 nm using UV/VIS spectrophotometer. The effects of scan temperature, light pre-gamma irradiation, dose rate, relative humidity and stability of the absorbance of solutions and films after irradiation were investigated. We found the dose sensitivity of TB solution and TB-PVA film dosimeters increases significantly with increases of the absorbed dose as well as with the increases of TB dye concentrations. The useful dose range of developed TB solutions and TB-PVA films dosimeters is in the range 0.125-1 kGy and of 5-20 kGy, respectively.

Keywords: Dose sensitivity; Radiochromic dosimeter; Thymol blue; Absorbance; Concentrations.

1. Introduction:
Dosimetry refers to the measurements of absorbed dose in material resulting from ionizing radiation such as x-rays, gamma ray and electron beam. Many types of dosimeters were introduced to meet the demands of new radiation processing of food irradiation, radiation sterilization and industrial applications. Radiochromic dosimeters are well-know dosimeters which contain specific type of dye in solution or binder such as polyvinyl alcohol (PVA) (Rabaeh et al, 2102; Kovács et al, 1996), polyvinyl buryral (PVB) (Mai et al, 2008; Buenfil- burgos et al, 1983) and Polystyrene (PS) (Kojima et al, 1997). These dosimeters exhibit
permanent change in color or color bleaching after exposed to ionizing radiation (Chan et al., 2017). The competition between various types of radio-chromic dosimeters depends on their accuracy, dose sensitivity and their response to different environmental conditions such as temperature, humidity, etc. (Basfer et al, 2014).

Polyvinyl butyral (PVB) films containing Thymol blue (TB) dye and chloral hydrate were investigated and found useful for ultraviolet radiation monitoring. (Abdel-Fattah et al. 2000). The color of TB-PVB films were changed from yellow to red after exposure to UV irradiation with a maximum sensitivity at 200nm. Various factors such as radiance exposure, irradiation wavelength, chloral hydrate concentration were also tested in this study. Recently, (Aldweri et al., 2017) introduced a novel solution and PVA film Radiochromic dosimeters based on Calcein dye for high dose gamma radiations. The results showed that the dose sensitivity in terms of change in color of these dosimeters increased significantly with increasing Calcein dye concentrations. In this study, Radio-chromic solution and film dosimeters containing different concentration of Thymol blue dye are introduced for high dose measurements in radiation processing applications. The dose range of developed TB solutions and TB-PVA films dosimeters is in the range 0.125-1 kGy and of 5-20 kGy, respectively.

2. Materials and methods

2.1. Preparation of TB solution and TB-PVA film dosimeters

The homogenous stock solution of Thymol blue (TB) dye was obtained from adding (0.018) g of TB dye (Sigma-aldrich, USA) in (300) ml double distilled water, and magnetically stirred at 25 °C temperature for 25 minutes. Two different concentrations of TB solution were
constructed from the previous stock solution as shown in Table 1. The TB solutions were poured in sealed 3 ml glass ampoules and protected from light at room temperature.

Table 1 Different concentrations of TB dye solution.

<table>
<thead>
<tr>
<th>Solution No.</th>
<th>Concentration (mM)</th>
</tr>
</thead>
<tbody>
<tr>
<td>1</td>
<td>0.129±0.0062</td>
</tr>
<tr>
<td>2</td>
<td>0.0645±0.0031</td>
</tr>
<tr>
<td>3</td>
<td>0.0323±0.0013</td>
</tr>
</tbody>
</table>

The homogeneous polyvinyl alcohol (PVA) solutions were obtained by adding 21.0 g PVA (Mw 50,000 g/M, Sigma-Aldrich, USA) in 300 ml of double distilled water. The mixture was heated to 70 °C and magnetically stirred for 20 hours until a complete homogenous solution was obtained. After decreasing the temperature to 25 °C, the solution was divided into 25 ml samples. Next, various concentrations of TB solution (see Table 1) were added to each 25 ml PVA solution and stirred for 20 hours until a complete homogenous TB PVA solution was obtained as demonstrated in Table 2. The solution was transfer into leveled glass plates (15x15 cm²) for 4 days to dry at 25 °C. Films were detached from the glass plate, cut into small pieces and dried at 25 °C for another 4 days. Finally, the TB-PVA films were wrapped with black mini envelopes to protect them from dust, light and moisture. The average thickness of TB-PVA films were about 290±10μm.
Table 2 Different concentrations of TB-PVA film dosimeter.

<table>
<thead>
<tr>
<th>Film No.</th>
<th>Concentration (mM)</th>
</tr>
</thead>
<tbody>
<tr>
<td>1</td>
<td>0.0645±0.0032</td>
</tr>
<tr>
<td>2</td>
<td>0.0513±0.0021</td>
</tr>
<tr>
<td>3</td>
<td>0.0320±0.0015</td>
</tr>
</tbody>
</table>

2.2. Gamma Irradiation:

Co-60 isotope source (Gamma-cell type PX-γ -30, Issledovatelj, Russia) with 1.25 MeV gamma beam was used to irradiate TB solutions and films dosimeters. Ferrous sulphate solution dosimeter was employed to calibrate the gamma cell, and the average dose rate of the gamma cell was about 0.48 kGy/h. The accuracy of TB dosimeters was investigated by irradiated a set of three samples for each absorbed dose. The dependence of TB-PVA films dose response on relative humidity (33.6 - 97.2%) during gamma irradiation has been conducted using three saturated salt solutions, namely K2SO4 (97.2%), NaCl (75.5%) and MgCl2×6H2O (33.6%) regarding to method described by (Levine et al, 1979). The TB-PVA films were stored in a given relative humidity for 72 hours before gamma irradiation to achieve stable environment (ASTM Standard Practice E2701-09, 2009).

2.3 Absorbance measurement:

Absorbance spectra of un-irradiated and irradiated TB solution as well as TB-PVA dosimeters were measured using UV-Vis spectrophotometer (Shimadzu, Japan). The optimum scanning wavelength range was determined from 400 - 650 nm and the absorbance of unirradiated and irradiated TB dosimeters were taken at 434 nm. Thermostated UV cary,
(Agilent, USA) was connected to UV-Vis spectrophotometer to investigate the effect of scanning temperature in the range from 0 to 80 °C.

3. Results and discussion

3.1. Effect of TB concentration on dose response of the solution dosimeters

The effect of the TB concentrations on the dose response of TB solutions was investigated by fabricating different compositions of TB solution dosimeters as shown in Table 1. The color bleaching of TB solution dosimeters increase gradually with increasing absorbed dose up to 0.125 kGy, then it tends to saturate up to 1 Gy (see Fig 1).

![Bleaching of thymol blue solution](image)

Fig. 1 Color bleaching of TB solution with increasing absorbed dose

The absorbance curves of TB solution dosimeters were established in terms of change in absorption peak measured at 434 nm. Figure 2 shows the absorbance spectra of TB solution (0.129 mM) as a function of wavelength.
Fig. 2 Absorption spectra of 0.129 mM TB solution dosimeter as a function of wavelength for various doses.

Absorbance of unirradiated and irradiated TB solution dosimeters with different concentrations of TB dye is demonstrated in Fig.3. The color bleaching of TB solution dosimeters increases significantly with increasing absorbed dose up to 1 kGy, which can be seen from decrease of the individual relative absorbance-dose curve. As the absorbed dose increases, more hydrate electrons and free radicals are generated leading to breakage of bonds of TB dye, resulting in the disappearance of chromophore. The accuracy of TB dosimeters was investigated by irradiated a set of three samples for each absorbed dose, but the dose variation between them was insignificant for all doses. The error bars in all figures were calculated from three different samples.
Fig 3 Absorbance of TB solution dosimeters containing different concentrations of TB as a function of absorbed dose (a) up to 4kGy, and (b) up to 0.75kGy.

The dose sensitivity was taken from the absolute slope of linear plot of Absorbance versus absorbed dose up to 0.75 kGy of Fig. 4. The results show that the dose sensitivity value increases gradually with increasing TB dye concentrations as shown in Fig. 4, leading to the increase of bleaching of TB solution dosimeters. Therefore, solution containing higher concentrations of TB dye is associated with higher dose dosimetry.

![Graph showing dose sensitivity vs. TB solution concentration](image)

Fig 4. The dose sensitivity for different TB solutions as a function of TB concentrations.

### 3.2 Pre and Post irradiation stability of TB solution dosimeters

The pre-irradiation stability is investigated by storing part of the 0.129 mM TB solution in the dark, and the other part under laboratory fluorescent light (100 W) at 1.5 meter distance from the light. The samples were kept under normal laboratory conditions of temperature and humidity. In the same way, the absorbance of the irradiated 0.129 mM TB solution (3 kGy) was measured every 24 hour using UV-Vis spectrophotometer for 7 days after irradiation. The results show no
change (less than 1% ;σ) in the absorbance of the solution up to 7 days (see Figure 5), which means that the TB solution has high stability, while TB solutions stored in light showed a decrease in optical absorbance with increase of storage time.

Fig. 5 Pre and post-irradiated 0.129 mM TB solution that stored in dark and in light as a function of storage time.

3.3. Effect of TB concentration on dose response of the TB-PVA film dosimeters

The effect of the TB concentrations on the dose response of TB-PVA films was investigated by fabricating different compositions of TB-PVA film dosimeters as shown in Table 2). The specific absorbance (absorbance per thickness in mm) curves of TB-PVA film dosimeters were established in terms of change in absorption peak measured at 434 nm per thickness in mm. The dose response of unirradiated and irradiated TB-PVA film dosimeters with different concentrations of TB dye is demonstrated in Fig.6. The color bleaching of TB-PVA film dosimeters increases significantly with increasing absorbed dose up to 30 kGy, which can
be seen from decrease of the individual relative specific absorbance-dose curve. As the absorbed dose increases, more hydrate electrons and free radicals are generated leading to breakage of bonds of TB dye, resulting in the disappearance of chromophore. The accuracy of TB Film dosimeters was investigated by irradiated a set of three films for each absorbed dose, but the dose variation between them was insignificant for all doses.

Fig 6 Specific absorbance of TB-PVA film dosimeter that containing different concentrations of TB as a function of absorbed dose.

The dose sensitivity was taken from the absolute slope of linear plot of Absorbance versus absorbed dose up to 30 kGy of Fig. 6. The results show that the sensitivity value increases gradually with increasing TB dye concentrations as shown in Fig. 7, leading to the increase of bleaching of TB solution dosimeters. Therefore, film containing higher concentrations of TB dye is associated with higher dose dosimetry.
3.4 Effect of scanning temperature on TB film dosimeters

The effect of scanning temperature on TB-PVA film was investigated using Thermostated UV cary. The specific absorbance curves of TB-PVA film dosimeters were created in terms of change in absorption peak measured at 434 nm for 0.0645 mM TB-PVA film, (see Fig. 8). A set of three samples was read for each scanning temperature and the average value for each set was reported. We found an insignificant change in dose responds of TB dosimeters with increases of scanning temperature, indicates that TB-PVA film dosimeters are independent upon the scanning temperature up to 80 °C.

Fig 7. The dose sensitivity for different TB film as a function of TB concentrations.
Fig. 8. Dose response of 0.0645 mM TB-PVA film at 15 kGy

3.5 Effect of relative humidity on the dose response of TB-PVA film dosimeters.

The effect of relative humidity on the TB-PVA film dosimeters was conducted by storing 0.0645 mM TB-PVA film in vials containing different relative humidity environments (i.e. in 34%, 74% and 97% relative humidity) for 3 days, then these films were irradiated to 5 and 10 kGy in the same vials. A set of three films was put into each vial, and the average specific absorbance was reported for each set. The dose response of the irradiated films as a function of relative humidity is shown in Figure 9. We found the specific absorbance changes slowly with increasing relative humidity, indicating that TB-PVA poses a good stability in this applicable range of relative humidity.
Fig. 9 Dose response of 0.0645 mM TB-PVA at 5 and 10 kGy as a function of relative humidity

3.6 Pre and Post irradiation stability of TB-PVA film dosimeters

The pre-irradiation stability is investigated by storing part of the 0.0645 mM TB-PVA films in the dark, and the other part under laboratory fluorescent lights (100 W) at 1.5 meter distance from the light. The samples were kept under normal laboratory conditions of temperature and humidity. In the same way, the absorbance of the irradiated 0.0645 mM TB-PVA (15 kGy) was measured every 24 hour using UV-Vis spectrophotometer for 7 days after irradiation. The results show no change (less than 1% ;σ) in the absorbance of the films up to 7 days (see Figure 10), which means that the TB-PVA film has high stability, while TB-PVA films stored in light showed a decrease in optical absorbance with increase of storage time.
Fig. 10. Pre and post-irradiated 0.0645 mM TB-PVA film that stored in dark and in light as a function of storage time.

4. Conclusions

Radiochromic solution and film dosimeters containing different concentrations of Thymol blue (TB) dye are introduced for dosimetry applications. The dose response in terms of change in absorbance of TB solutions and film dosimeters increases considerably with increase of absorbed dose. The dose sensitivity value of TB solution and film dosimeters increases gradually with increasing TB dye concentrations. The results show that an insignificant change in dose responds with increases of scanning temperature, therefore TB-PVA film dosimeters are independent upon the scanning temperature up to 80 °C. The specific absorbance changes slowly with increasing relative humidity, indicating that TB-PVA poses a good stability in this applicable range of relative humidity.
Acknowledgment

The authors would like to acknowledge the support of Hashemite University (HU 50-5-2014) in funding this project.

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Fig. 4 The dose sensitivity for different TB solutions as a function of TB concentrations.

Fig. 5 Pre and post-irradiated 0.129 mM TB solution that stored in dark and in light as a function of storage time.

Fig. 6 Specific absorbance of TB-PVA film dosimeter that containing different concentrations of TB as a function of absorbed dose.

Fig. 7 The dose sensitivity for different TB film as a function of TB concentrations.

Fig. 8 Dose response of 0.0645 mM TB-PVA film at 15 kGy

Fig. 9 Dose response of 0.0645 mM TB-PVA at 5 and 10 kGy as a function of relative humidity

Fig. 10 Pre and post-irradiated 0.0645 mM TB-PVA film that stored in dark and in light as a function of storage time.
Highlights

- Radiochromic dosimeter based on Thymol blue dosimeter.
- Linear dose response range up to 30 kGy.
- The colour bleaching of Thymol blue increases gradually with increasing dose.