Nuclear Engineering and Technology 55 (2023) 3383–3387

Contents lists available at ScienceDirect

Nuclear Engineering and Technology

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

Original Article

Novel polyvinyl alcohol film dosimeter containing 3-(4,5-Dimethylthiazol-2-yl)-2,5-diphenyltetrazolium bromide dye for high dose application

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A R T I C L E I N F O

Article history:
Received 20 September 2022
Received in revised form
6 May 2023
Accepted 5 June 2023
Available online 14 July 2023

Keywords:
Gamma radiation
Radio-chromic film
Optical absorbance
Polyvinyl alcohol
3-(4,5-Dimethylthiazol-2-yl)-2,5-diphenyltetrazolium bromide
Formazan

A B S T R A C T

A new dyed polyvinyl alcohol (PVA) film dosimeter based on 3-(4,5-Dimethylthiazol-2-yl)-2,5-diphenyltetrazolium bromide (MMT) tetrazolium dye is proposed in this study for measuring high gamma radiation dose. Gamma cell irradiator that contains Co-60 gamma-ray source was used to expose the novel MMT-PVA films to different doses up to 25 kGy. The changed in optical property of irradiated and unirradiated films were characterized by UV–Vis spectrophotometer. The results show that the dose sensitive and the linear range of irradiated films were increased considerably with increase of MMT concentration from 1 to 5 mM. The dose response of dyed PVA film changed substantially with changing relative humidity (12–74%) as well as irradiation temperature (10–40 °C). The absorbance of the unirradiated films does not change up to 10 days in dark while a significant increase in their absorbance was reported for similar films under fluorescent light. The irradiated dosimeters that kept in dark showed a perfect stability for 54 days. It was found that no obvious impact of dose rate on the irradiated MMT-PVA film dosimeters.

1. Introduction

Dosimeters are now essential for determining the dose of ionizing radiation exposed in one-dimensional, two-dimensional, or three-dimensional forms. As a result, it is critical to guarantee that the radiation dose is correct. The primary purpose of any dosimeter is to ensure long-term stability, simplicity, ease of use, and cost-effectiveness [1–6].

There are two main types of dosimeters: physical and chemical, depending on the irradiation effect on the material. Physical dosimeters which convert radiation energy into heat that can be precisely measured, such as calorimeters. They are primary dosimeters since they do not normally need to be calibrated with another radiation measuring instrument or solid-state dosimeters [7–10]. On the other hand, chemical dosimeters can be aqueous liquid chemical systems, such as the ferrous sulfate dosimeter, which is based on the oxidation of ferrous ions to ferric ions and is commonly used in radiation processing for calibration [11].

Polymeric dyed flexible films are the most well-known and inexpensive type of dosimeter, which are used in dose labels and indicators in radiation processing for routine dose monitoring. These films absorb the dose, resulting in a visual alteration in the form of a color shift in the visible region [12–14], as well as physical and chemical changes [15,16]. One of the most important requirements for dye and polymer selection in the production of dosimeters is a long-lasting color change [17–19]. Several dyes were tested such as dithizone dye [20,21], methyl red [22–24], calcien [25], leucomalachite green [26], thymol blue [27], and basic violet [28]. The main concepts being studied are the dye concentration and its effect on the dosimeter's response to the radiation specifications, stability, and storing conditions like humidity.

Tetrazolium salts are stable compounds that are soluble in water and alcohol that are quaternary derivatives of tetrazoles and therefore contain a ring of one carbon and four nitrogen atoms. Their solutions are colorless or very pale yellow, and the radiolytic reduction of these tetrazolium salts by hydrated electrons or

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https://doi.org/10.1016/j.net.2023.06.011

hydroxyalkyl radicals results in a ring opening and the production of colored water-insoluble compounds known as formazans [29,30]. Based on all these characteristics of tetrazolium salt, several scientists synthesized different types of film dosimeters and investigated their characteristics and behaviors under irradiation, including [31–36].

Polyvinyl alcohol (PVA) is frequently used for making polymeric films due to its good chemical and physical properties, excellent film-forming nature, and hydrophilicity, which makes it a highly permeable material. It is a water-soluble material containing significant numbers of hydroxyl groups and can be blended with the dye to obtain optical materials. It is used by many scientists with different types of dyes, like Rabaeh and his coworkers [37] who investigated the nitro blue tetrazolium (NBT) PVA films which irradiated with doses up to 50 kGy, and the dose sensitivity was increasing significantly when the increasing NBT dye concentration in other hand these films were sensitive to humidity and irradiation temperature. The same polymer, PVA, were used with different dye, which was methyl thymol blue (MTB) but with lower dose range, 2.5–20 kGy, which also showed a direct relation between the dye concentration and the sensitivity [38]. Al Zahrany and his group tested another type of dye with PVA, which is methyl red (MR), and was used for high dose range, up to 60 kGy, and they reported an enhancement when MR dye increase [24].

The current study introduces a radiochromic film dosimeter containing a PVA and a 3-(4,5-Dimethylthiazol-2-yl)-2,5-diphenyltetrazolium bromide (MMT) tetrazolium dye. The films were tested under high dose gamma radiation with analysis for many properties using absorbance values measured after each variable condition to determine how these film dosimeters could be used as a new radiochromic film dosimeter for measuring ionizing absorbed.

2. Materials and methods

2.1. Preparation of polymer film dosimeter

The fabrication process of Polyvinyl alcohol (PVA) films is set up by dissolving 7.2 g of PVA powder (Mw = 108.000 g/M, Polysciences Inc., USA) in 90 ml of distilled water at a temperature of 80 °C. This solution was magnetically stirred at the same temperature for 4 h and then left to cool down at room temperature. Following that, the PVA solution was divided into three parts for dissolving 3-(4,5-Dimethylthiazol-2-yl)-2,5-diphenyltetrazolium bromide (MMT) at three different concentrations: 1, 3, and 5 mM. Mixtures were stirred continuously for 24 h to acquire a homogeneously dyed PVA solution. MMT-PVA solutions were poured onto high-leveled horizontal glass plates and dried at room temperature for about 72 h. These films have been peeled and cut into 1 × 3 cm pieces, dried, stored, and prepared for irradiation. When the weight of the films is constant, the drying process is complete. By placing the films in a small paper envelop and sealing them in black plastic tape, they were protected from sunlight, fluorescent light, moisture, and dust. The film has a thickness of 60 ± 3 μm and is quite homogeneous.

Some samples with 5 mM MMT-PVA was stored in airtight vials that half filled with saturated salts (LiCl, $MgCl_2 \times 6H_2O$, $Mg(NO_3)_2 \times 6H_2O$ and NaCl) to obtain different environmental relative humidity of (12, 34, 55 and 75%, respectively) according to the method delivered by Levine et al. (1997). The samples were kept in the same environment for three days before irradiation to establish proper equilibrium and to study the effect of humidity on its reaction [39].

2.2. Irradiation

Gamma cell contains ^{60}Co gamma-ray source (Model GC-220 supplied by MDS Nordion, Canada) was used to irradiate the novel MMT-PVA films to different doses up to 25 kGy. The dose rate and energy of the source were 8.6 kGy/h and 1.25 MeV, respectively. Three samples were represented for each irradiation dose, and the median value was taken.

Gamma cell was connected to an air chiller system (Turbo-Jet, Kinetics, USA) to study the effect of temperature during irradiation. The gamma source was calibrated previously by Fricke solution dosimeter [40].

In addition of gamma ray beam irradiation, MMT-PVA films were also irradiated by mega voltage electron beam using electron beam accelerator (EBA) manufactured by (L3 Communications Ltd., USA) available at (Sure Beam Middle East Corp., Saudi Arabia) at 1 kGy/s dose rate and 10 MeV beam energy. The EBA was calibrated periodically by alanine film dosimeters that could be read out by electron spin resonance (Bruker Biospin, Model EPR, Germany).

2.3. Optical measurements

The radiation changed the absorbance property of these films, which were measured at 560 nm with a UV–Vis spectrophotometer (Model Lambda 850, PerkinElmer, USA). For each response measurement, three samples were read out and the median value has been recorded in our results, where the standard deviation value was less than 3%. Also, the dose response curves were established in terms of change in absorption peak measured at 560 nm per thickness in mm ΔA ($\Delta A = A_x - A_0$) versus the absorbed dose, where A_x and A_0 are absorbance values at 560 nm for irradiated and un-irradiated films (i.e for each dose (2.5, 5, 10, 15, 20 and 25), one film was read out (A_0), then the same film was irradiated to specific dose and read out again (A_x) and ΔA was obtained).

3. Results and discussion

3.1. MMT concentration effect

The effect of MMT dye concentration on the response of the novel MMT-PVA dosimeter films was studied using three concentrations of MMT (1, 3 and 5 mM). The dose response of these films in terms of specific absorbance (Absorbance per thickness in mm) is shown in Fig. 1 depicting a direct relation between the specific

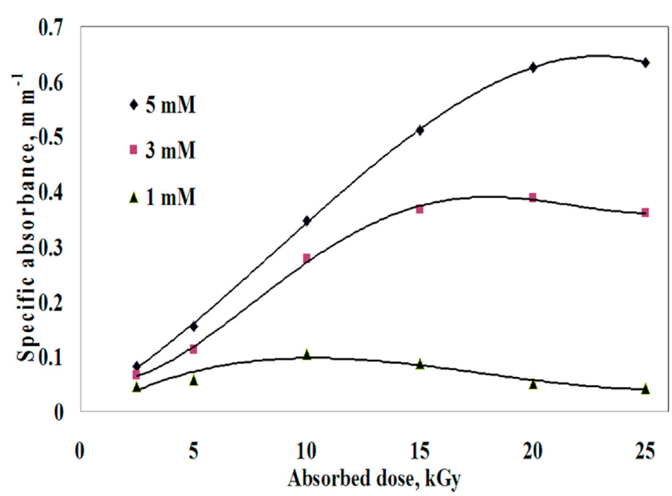

Fig. 1. MMT-PVA film dosimeter specific absorbance as a function of absorbed dose, with three different MMT concentrations, where the temperature and humidity in irradiation were about 20 °C and 55% respectively.

absorbance of MMT-PVA film and the absorbed dose. Also, when the concentration of MMT increases, the composition response becomes more sensitive to radiation and the linear range increases significantly. The radiation increases the number of hydrated electrons and free radicals produced, which stimulates the formation of colorful formazan by breaking N—N+ bonds. Indicating that MMT-PVA dosimeter films with higher MMT dye concentrations are more suited for high dosage applications. This result is consistent with the findings of other study that used PVA binder and NBT dye (Rabaeh et al., 2012) and it improved remarkably with increasing dye concertation.

3.2. The effect of irradiation temperature

To study the irradiation temperature effect on the MMT-PVA film response, the optimum composition with 5 mM MMT was irradiated to 10 and 20 kGy at four different temperatures, in the range of 10–40 °C, for each absorbed dose. A set of three films was used for each selected temperature and the average values are plotted in Fig. 2. After normalizing the variation in absorbance with respect to the irradiation temperature at 10 °C. Findings indicate that MMT-PVA films are sensitive to irradiation temperature. Therefore, the response of films has to be corrected under actual processing conditions [41].

The impact of irradiation temperature was also reported for PVA with NBT dye, which was studied by Rabaeh and his coauthors [37]. On the other hand, many previous film dosimeters like Red-perspex, Amber-perspex, FWT-60, and CTA FTR-125 films showed a significant effect of irradiation temperature on their performance [42].

3.3. The effect of humidity

The humidity impact on MMT-PVA films response has been studied by storing samples with 5 mM MMT in vials at various humidity levels (12%, 34%, 55% and 74% relative humidity) for three days, then the films were irradiated in the same vials to 10 and 20 kGy. A set of three films was used for every vial. The variation in absorbance of the irradiated films was normalized with respect to 12% relative humidity and plotted in Fig. 3. Results demonstrate that the dose response of MMT-PVA film increases considerably when the relative humidity increases. This agrees well with previous studies on dosimeters with dyes like NBT-PVA [37], and MR-PVA [24].

3.4. The stability

The stability of novel MMT-PVA film was assessed using MMT-PVA with 5 mM of MMT sample. Fig. 4 shows the absorbance measured up to 54 days of MMT-PVA films with 5 mM MMT that

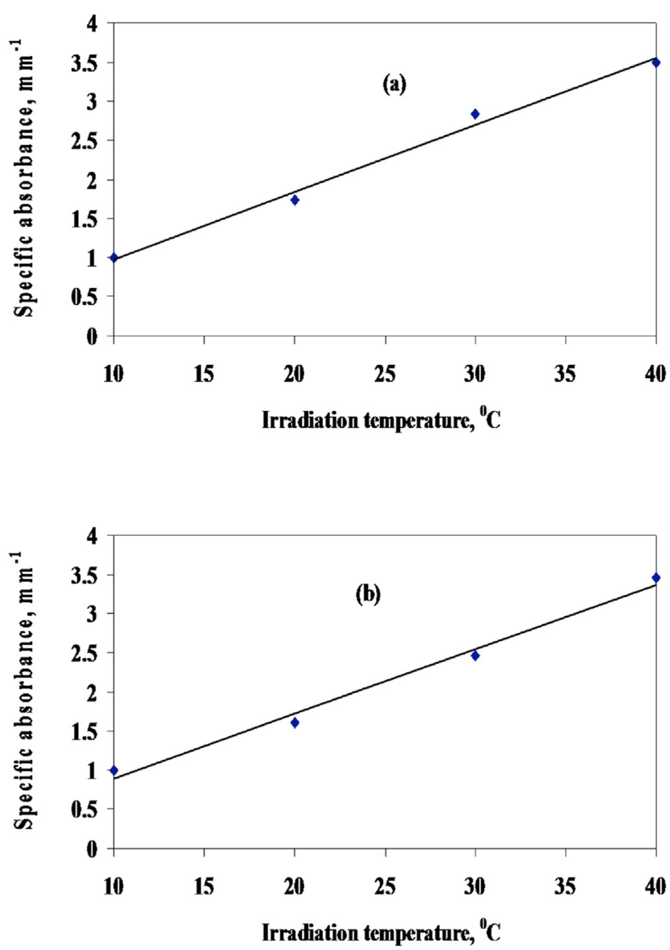

Fig. 2. Specific absorbance of 5 mM MMT-PVA film dosimeters normalized with respect to that at 10 °C versus irradiation temperature for (a) 10 kGy and (b) 20 kGy, where the humidity in irradiation was about 55%.

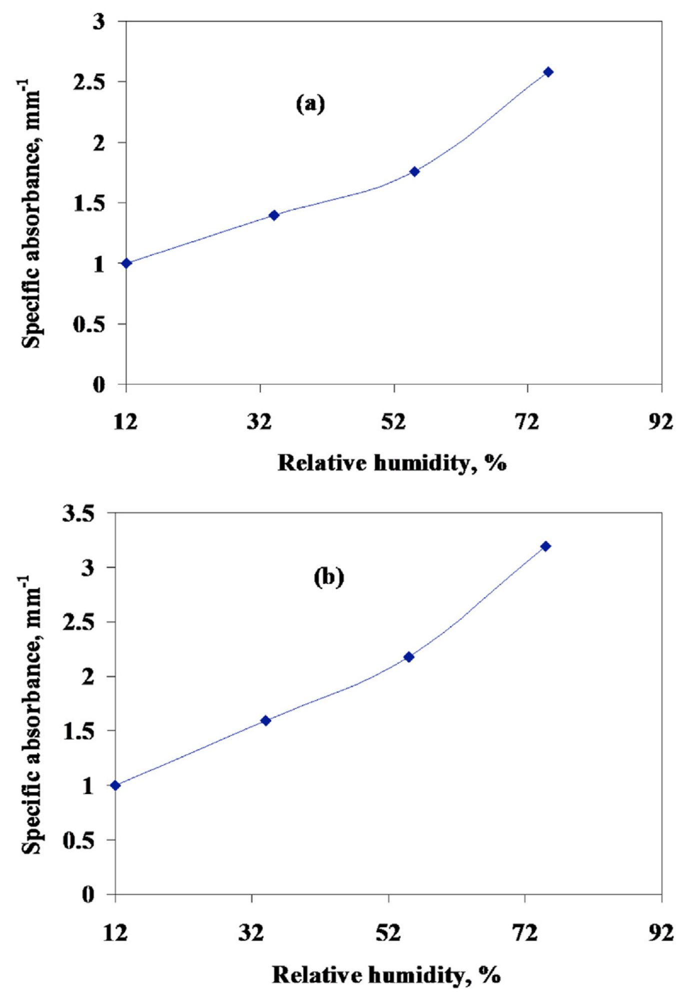

Fig. 3. Specific absorbance of 5 mM MMT-PVA film dosimeters at (a) 10 kGy and (b) 20 kGy that normalized to that at 12% relative humidity where the temperature in irradiation was about 20 °C.

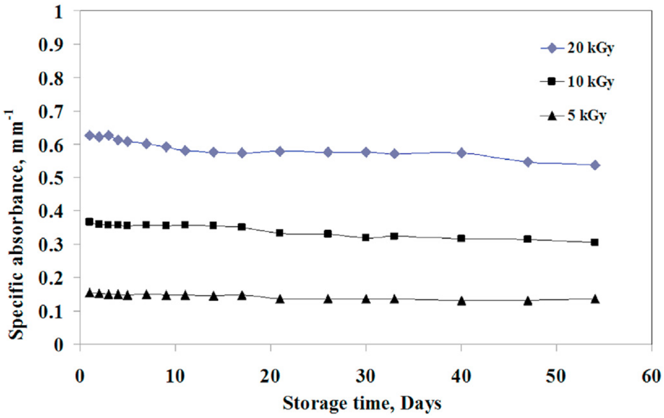

Fig. 4. Specific absorbance of irradiated 5 mM MMT-PVA film dosimeters for various doses absorbed where the temperature and humidity in irradiation were about 20 °C and 55% respectively.

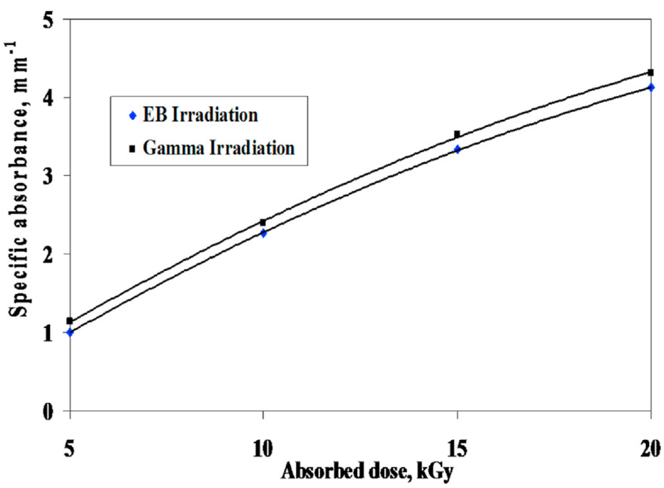

Fig. 5. Specific absorbance of 5 mM MMT-PVA film dosimeters normalized with respect to that at 5 Gy exposed to two types of irradiation beam at varied absorbed doses, where the temperature and humidity in irradiation were about 20 °C and 55%.

were irradiated in three doses, 5, 10, and 20 kGy, and kept in the dark under normal laboratory conditions, temperature, and humidity. A set of three films was used for each dose. Findings indicate no remarkable change (less than 5%; 1σ for MMT-PVA) in the specific absorbance of the PVA up to 54, and it appears more stable as the dose decreases. This conclusion coincides with prior work by Rabaeh and his group, who tested the stability of NBT-PVA films for up to 30 days and found a good stability, particularly on samples irradiated with lower doses [37].

3.5. The effect of dose rate

The impact of dose rate on the MMT-PVA film absorbance was investigated using two types of radiation; a 1.25 MeV gamma-ray from a^{60}Co source at a mean dose rate of 8.6 kGy/h and an electron beam accelerator (SureBeam Middleeast Co. Saudi Arabia) with dose rate of 1 kGy/s. This property was tested in four different absorbed doses: 10, 20, 30, and 40 kGy. The results are depicted in Fig. 5 shows no discernible influence of dose rate on MMT-PVA film dosimeters, which is consistent with [33].

4. Conclusions

Novel polyvinyl alcohol (PVA) film dosimeter with different concentration of 3-(4,5-Dimethylthiazol-2-yl)-2,5 diphenyltetrazolium bromide (MMT) dye was introduced in this work for radiation processing. As a result of increasing the dye concentration from 1 to 5 mM, the dose sensitivity of PVA-MMT film was increased remarkably, moreover, the linear dose range increased also significantly. The results showed the PVA-MMT film should be protected from fluorescent light and the film should be corrected under actual processing conditions that determined the exact values for relative humidity and irradiation temperature. The results show that the specific absorbance of the un-irradiated films does not change (less than 6%; 1 σ) after 10 days. No apparent influence of dose rate on MMT-PVA film dosimeters was reported. The results show this low cost film can be used in similar manner to many types of radio-chromic dosimeters that are commercially available such as Gafchromic™ EBT, Risø B3, and Red Perspex [43,44]. In future work, we will focus on using different binder such as polyvinyl butyral to reduce the effect of relative humidity during irradiation.

Declaration of competing interest

Potential conflicts don't exist.

Acknowledgments

This work was supported by King Abdulaziz City for Science and Technology (KACST), KSA (grant 10-BIO960-20).

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