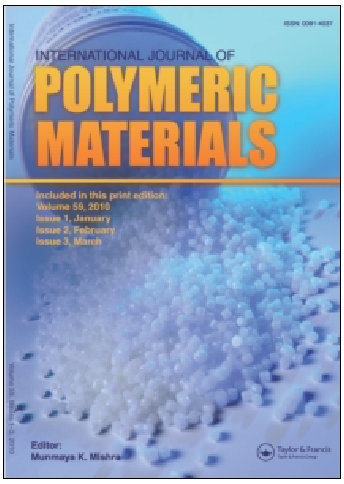

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THIN FILM DOSIMETERS BASED ON ROSE BENGAL DYED POLY(VINYL ALCOHOL)

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Dyed poly(vinyl alcohol) (PVA) films were prepared by casting the aqueous solutions of PVA incorporating Rose Bengal (RB) dye with or without chloral hydrate on a horizontal glass plate. Such films are useful as routine high-dose dosimeters. These flexible plastic film dosimeters are bleached when exposed to γ-ray photons. The useful range of application of these films depends on the concentration of chloral hydrate. RB/PVA film without chloral hydrate is suitable for application in the range of absorbed doses 10–150 kGy. The maximum range of RB/PVA films containing chloral hydrate ranges from 5 to 20 kGy depending on, and inversely proportional to, chloral hydrate concentration in the film. The radiation-chemical yield (G-value) of films was calculated to be in the range from 0.272 to 10.3 μmol/J, where it increases with the increase of both RB and chloral hydrate concentration in the film. The effects of temperature and relative humidity during irradiation as well as pre- and post-irradiation storage, on the radiation response of films are studied. Although, the response of these films depends on relative humidity during irradiation, they are highly stable for long times before and after irradiation when stored in dark and they are not affected by temperature that ranged from 0°C to 45°C during irradiation.

Keywords: rose bengal, poly(vinyl alcohol), radiation dosimetry, thin polymeric films

INTRODUCTION

Thin polymeric film dosimeters are the most commonly used indicators and monitors of absorbed dose for routine use in radiation processing by electron beam and γ-rays [1–7]. Many dyed poly(vinyl alcohol) films have been developed and investigated for the possibility of their being used to measure neutron doses in nuclear reactors and doses of X-rays, γ-rays and electron beams [8–15]. All these dyed PVA systems are bleached by irradiation, the extent being to which the color change is used for

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determining the absorbed dose. Based on the idea of mixing, in poly(vinyl alcohol), two dyes having different sensitivities to radiation, a new label dosimetry system has been developed [16]. A new radiation-sensitive indicator consisting of poly(vinyl alcohol) film containing pH-indicating dye and a water-soluble chlorine-containing substance has been developed by Abdel-Fattah *et al.* (1996) [15]. Chloral hydrate was previously used in polymeric film dosimeters as an acid-releasing agent [17].

In the current work, thin films of PVA colored with Rose Bengal dye (RB) in absence or presence of chloral hydrate as a radiation sensitizer are investigated to be used as dosimeters for radiation processing. The effects of temperature and relative humidity during irradiation as well as pre- and post-irradiation stability on the response of the film are discussed.

EXPERIMENTAL

Preparation of Stock Solution of RB Dye

The stock solution of RB dye was prepared by dissolving 0.08 g of Rose Bengal dye (Product of ALDRICH) in 50-mL bi-distilled water.

Preparation of RB/PVA Films

12 grams of fully hydrolyzed poly(vinyl alcohol) powder (99 – 100%) from J.T. Baker Chemicals Co., U.S.A. were dissolved in 200 mL bi-distilled water at about 60°C. The solution was kept well stirred at the same temperature for about 48 h; then left to cool. To each 25 mL of the well-mixed solution, 3 mL of stock solution of RB and 0, 0.03, 0.06, 0.1, 0.15 or 0.2 g of chloral hydrate (product of Merck, Germany) were added, stirred, casted on a 15 × 15 cm horizontal glass plate and dried at room temperature for about 48 h. Six films were obtained containing the same concentration of RB (0.32 phr[1]) and different concentrations of chioral hydrate (0, 2, 4, 6.7, 10 and 13.3 phr). Other two films were prepared containing the same concentration of chloral hydrate (10 phr) and different concentrations of RB (0.107 and 0.214 phr). The films thus obtained of thickness 0.06 ± 0.003 mm (1σ), were then cut into pieces (1 × 1 cm) and stored for the purpose of this study.

Apparatus

Irradiations were carried out with gamma radiation in the ^{60}Co gamma chamber 4000A (product of India). The absorbed dose rate in the irradiation facility was measured to be 3.0 kGy/h, using Fricke dosimetry [$G(Fe^{3+}) = 1.62\ \mu mol/J$] [18]. Five films at each dose are grouped together,

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[1] phr = part per hundred parts by weight of resin.

sandwiched between two PMMA plates of 3 mm thickness to maintain electronic equilibrium conditions. Before irradiation, the films and the plates were preconditioned at 33% relative humidity for 24 h in a glass box, enclosed in and sealed by aluminum–polyethylene laminate foil, then irradiated at the central spatial position of the sample chamber using a specially designed holder made from polystyrene. The temperature during irradiation was ca 30°C.

Uvikon 860 spectrophotometer (product of KONTRON Co. Ltd., Switzerland) was used to measure the absorption spectra of the unirradiated and irradiated films. The film thickness was measured using a Digitrix-Mark II thickness gauge (precision ± 1 μm, 1σ).

RESULTS AND DISCUSSION

Absorption Spectra

The absorption spectra of the RB/PVA film without chloral hydrate ([RB] = 0.32 phr) were recorded before and after irradiation to different doses, and are shown in Figure 1. The absorption spectrum of the

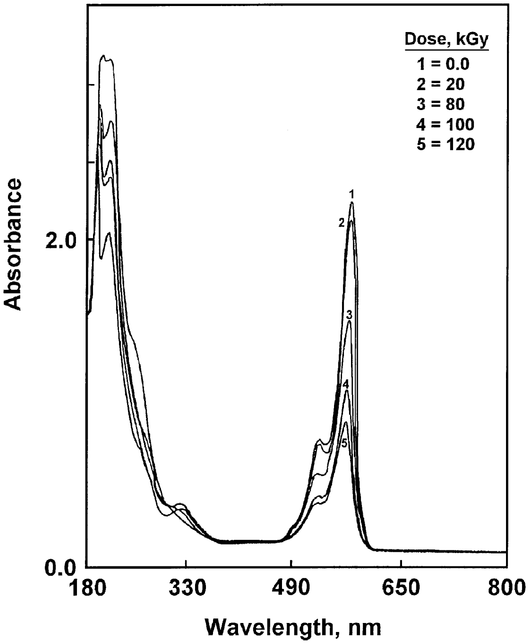

FIGURE 1 The absorption spectra of RB/PVA films (without chloral hydrate) unirradiated and irradiated to different absorbed doses. [RB] = 0.32 phr.

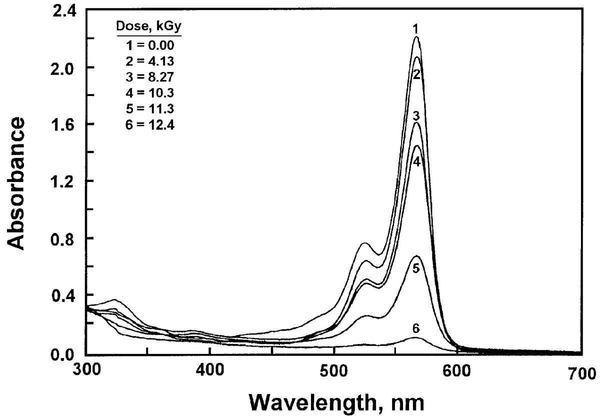

FIGURE 2 The absorption spectra of RB/PVA films unirradiated and irradiated to different absorbed doses. [RB] = 0.32 phr; [chloral hydrate] = 10 phr.

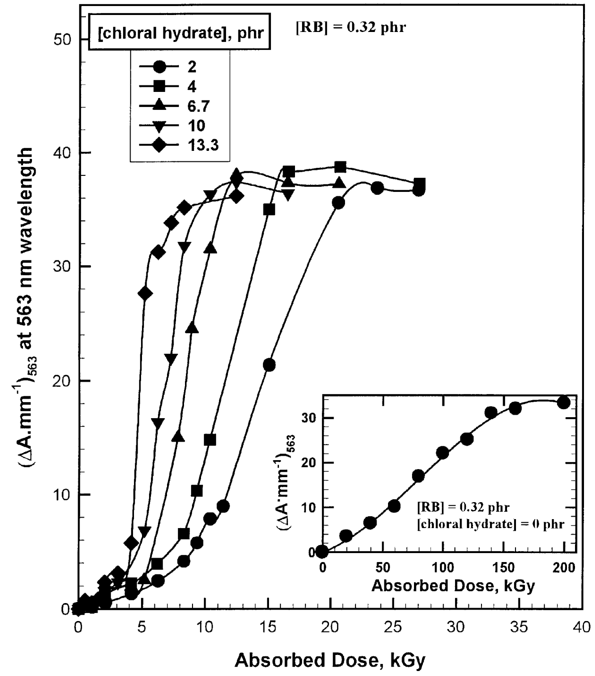

FIGURE 3 Change of $\Delta A \cdot \mathrm{mm}^{-1}$ as a function of absorbed dose of RB/PVA films containing 0.32 phr of RB and different concentrations of chloral hydrate. $\lambda_{\max} = 563$ nm.

unirradiated RB/PVA film shows a main absorption band in the visible region peaking at 563 nm, which is characteristic of rose color (Fig. 1, curve 1). The amplitude of this peak at 563 nm decreases gradually with the increase of dose of γ-ray photons.

Figure 2 shows the absorption spectra of RB/PVA films containing chloral hydrate ([chloral hydrate] = 10 phr; [RB] = 0.32 phr) before and after irradiation to different γ-doses. It can be noted that in the films containing chloral hydrate, there is a very fast radiation-induced bleaching of the rose color of the films. In other words, the presence of chloral hydrate in the films increases the rate of bleaching of RB dye, *i.e.*, chloral hydrate acts as a sensitizer.

Effect of Chloral Hydrate Concentration on Response

Figure 3 shows the effect of chloral hydrate concentration on the response of PVA films containing 0, 2, 4, 6.7, 10 or 13.3 phr of chloral hydrate at fixed concentration of RB (0.32 phr) and irradiated different doses. The insert in Figure 3, shows the response curve of RB/PVA film without chloral hydrate.

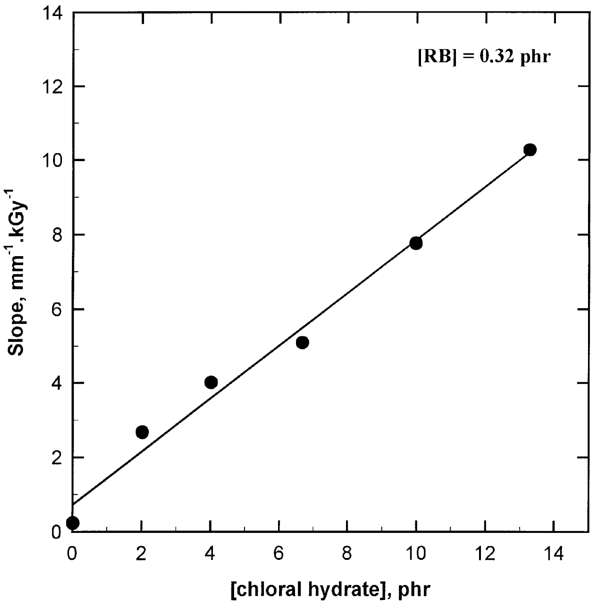

FIGURE 4 Change of response, slope of straight sections of curves given in Figure 3, as a function of the concentration of chloral hydrate in the films.

The response curves were established in terms of change in optical density at 563 nm per unit thickness, $\Delta A \cdot \mathrm{mm}^{-1}$, against the absorbed dose ($\Delta A = A_o - A_i$, where A_o and A_i are values of optical absorbance at 563 nm for unirradiated and irradiated films, respectively). It can be noticed that all curves show the same trend, they have S-shape and tend to saturate at higher doses, but all curves differ in their response value (slope of the straight part in the curve). Figure 4 shows the relation between the slope of the straight parts of curves in Figure 3 and the concentration of chloral hydrate. It can be seen that the slope increases linearly with the increase of chloral hydrate concentration.

The curves in Figure 3 reach saturation at different doses, depending on the concentration of chloral hydrate in the film. Figure 5 shows the relation between the dose at saturation and the concentration of chloral hydrate. It can be seen that, the dose at saturation decreases exponentially with the increase of chloral hydrate concentration. The insert in Figure 5 shows the same relation on a log–log scale. These results reflect the pronounced sensitizing effect of chloral hydrate on the radiation-induced bleaching of rose Bengal. The maximum range of dose varies from 5 to 150 kGy depending on the concentration of chloral hydrate in the film. From this

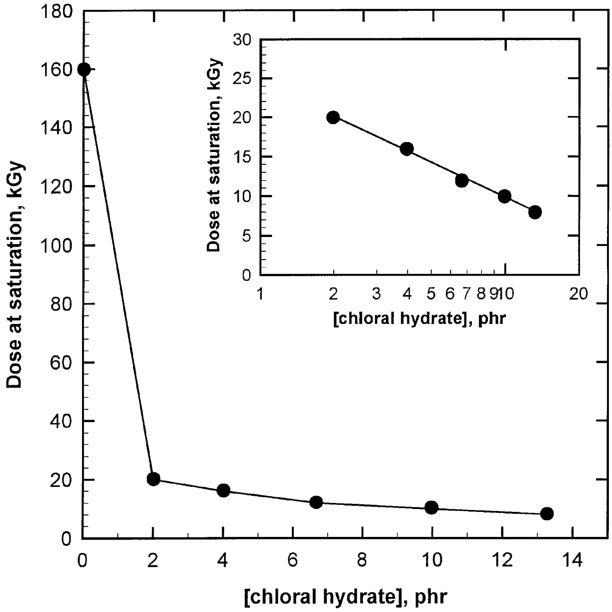

FIGURE 5 Variation of dose at saturation of RB/PVA films containing 0.32 phr of RB as a function of concentration of chloral hydrate. The insert gives the same relation on a log–log scale.

figure one can predict the suitable concentration of chloral hydrate in a film for a specific range of application.

Effect of RB Concentration on Response

Figure 6 shows the response curves of RB/PVA using different concentrations of RB (0.107, 0.213, or 0.32 phr) at constant concentration of chloral hydrate (10 phr). All curves show the same trend in which they have S-shape, but reach saturation at different doses depending on concentration of RB. Figure 7 shows the relation between dose at saturation and concentration of RB. The dose at saturation increases linearly with the increase of RB concentration.

Radiation-Chemical Yield

The radiation-chemical yield (G-value) is defined as the number of moles of dye degraded by the absorption of 1 J of energy (unit: mol/J). The G-value is calculated from the general relation [19] as follows:

$$G(\text{-dye}) = \frac{\Delta A}{D \cdot \varepsilon \cdot \rho \cdot b}\ \text{mol/J}$$

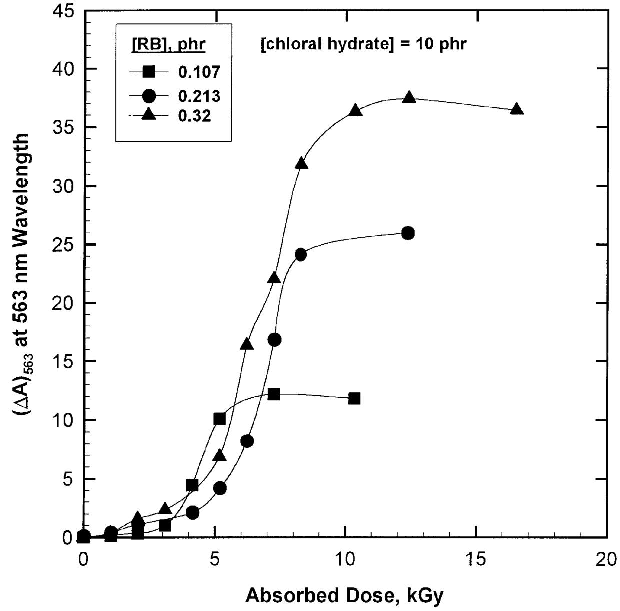

FIGURE 6 Change of $\Delta A \cdot \text{mm}^{-1}$ as a function of absorbed dose of RB/PVA films containing 10 phr of chloral hydrate and different concentrations of RB. $\lambda_{\text{max}} = 563\,\text{nm}$.

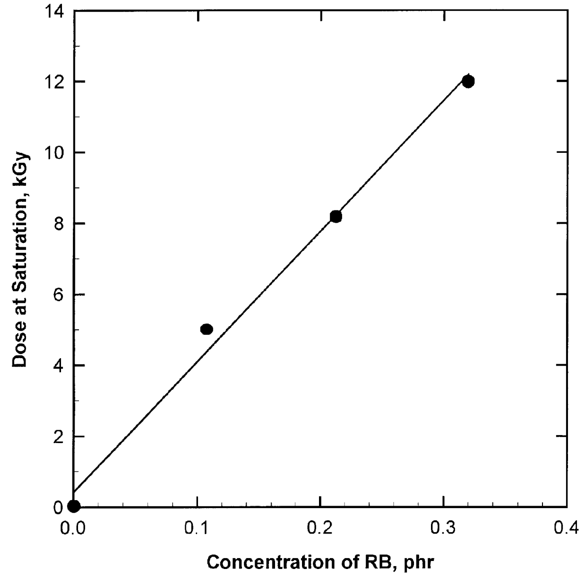

FIGURE 7 Variation of dose at saturation of RB/PVA films containing 10 phr of chloral hydrate as a function of concentration of RB.

where

ΔA = the change in the absorbance at λ_{max}.
b = the optical bath length (cm).
ε = the linear molar extinction coefficient at λ_{max} ($L \cdot mol^{-1} \cdot cm^{-1}$).
ρ = the density of the film ($g \cdot cm^{-3}$).
D = the absorbed dose (Gy).

Figure 8 shows the relation between A_o/b (cm^{-1}) as a function of concentration of rose Bengal (in mol/L) giving a straight line; its slope is the molar extinction coefficient, ε, and it was found to be 97954.2 $L \cdot mol^{-1} \cdot cm^{-1}$. Using the density of PVA ($1.25\,g \cdot cm^{-3}$), the G(-RB) was calculated for different films containing different concentrations of rose Bengal and chloral hydrate and the results are tabulated in Table 1. The change of G-value as a function of concentration of chloral hydrate at the same concentration of rose Bengal (0.32 phr) is shown in Figure 9(a). It can be seen that G-value increases gradually with the increase of concentration of chloral hydrate up to about 7 phr and increases dramatically at concentrations of chloral hydrate higher than 7 phr. Figure 9(b) shows the relation between G(-RB) and concentration of rose Bengal at the same concentration of chloral hydrate (10 phr). It was found that, G(-RB) increases gradually with

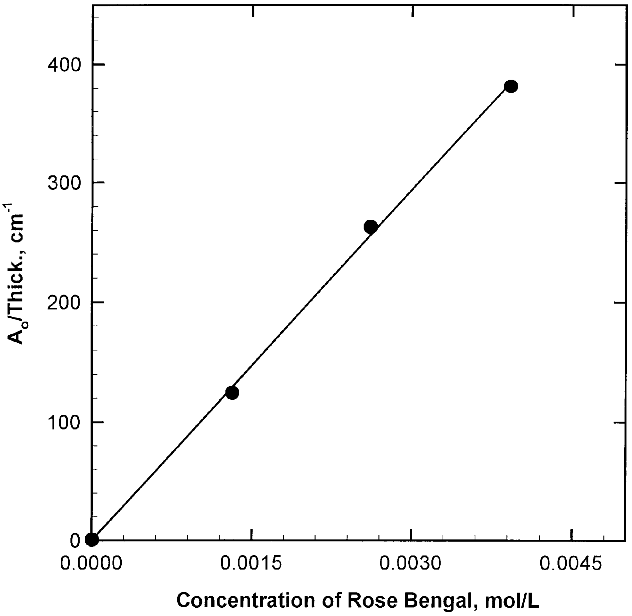

FIGURE 8 The relation between A_o/Thick., as a function of concentration of RB.

TABLE 1 *G*-value of PVA/RB films at different concentrations of chloral hydrate and rose Bengal

Film composition		*G-value,*
[*chloral hydrate*], *phr*	[*rose Bengal*], *phr*	*μmol/J*
0	0.32	0.272
2	0.32	0.82
4	0.32	1.23
6.7	0.32	1.58
10	0.107	4.393
10	0.213	6.664
10	0.32	7.78
13.3	0.32	10.279

the increase of concentration of rose Bengal and tends to saturate at higher concentrations.

Effect of Humidity during Irradiation

The effect of relative humidity (RH) during irradiation on response of RB/PVA film ([chloral hydrate] = 10 phr, [RB] = 0.32 phr) was investigated by

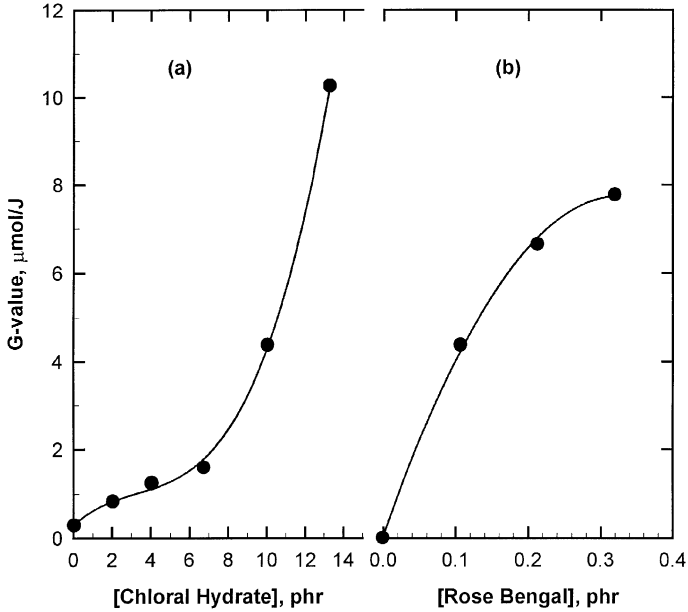

FIGURE 9 (a) *G*-value as a function of chloral hydrate concentration. [RB] = 0.32 phr. (b) *G*-value as a function of RB concentration. [chloral hydrate] = 10 phr.

irradiating the films to a dose of 6.2 kGy at different relative humidities (0, 12, 33, 54, 76 and 92%). Irradiation was carried out while the films were suspended over various saturated-salt solutions in an enclosed jar [20], except for the 0% RH, which was made with films suspended over dried silica gel. The films were stored before irradiation for a 48-h period under the same RH conditions as when irradiated, so that equilibrium moisture content in the dosimeter could be established during irradiation. Figure 10 shows the variation in response ($\Delta A \cdot \mathrm{mm}^{-1}$) as a function of percentage relative humidity during irradiation relative to that at 33%. It can be seen that the response increases gradually with the increase of RH% and it reaches to the maximum at 33% then decreases gradually with increasing RH%. For dosimetry, it may be possible to reduce the influence of humidity by preconditioning of the films at about 33% RH and sealing them in vapour-tight pouches before irradiation or, if that is impractical, one should maintain conditions of calibration as close as possible to the conditions of use.

Effect of Temperature during Irradiation

The temperature during irradiation was investigated by irradiating RB/PVA films ([RB] = 0.32 phr and [chloral hydrate] = 10 phr) to a dose of 6.2 kGy at

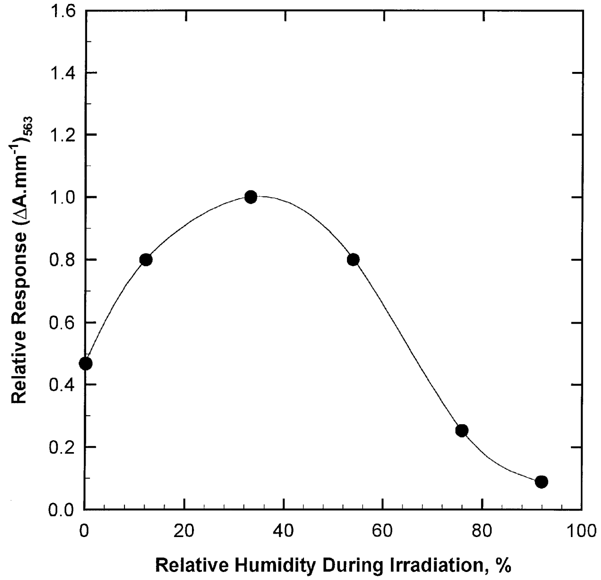

FIGURE 10 Variation of relative response of RB/PVA films ($\Delta A \cdot \text{mm}^{-1}$, at 563 nm) as a function of relative humidity during irradiation to a dose of 6.2 kGy. [RB] = 0.32 phr; [chloral hydrate] = 10 phr.

different temperatures (0, 25, 40, 50, 60 and 70°C) using liquid thermal baths [21]. The variation in response, $\Delta A \cdot \text{mm}^{-1}$, as a function of temperature during irradiation is shown in Figure 11. It can be seen that there is no appreciable effect of temperature on response in the range from 0 to ~45°C, where the variation of response in this range is less than 5%. At temperatures higher than 45°C, the response increases gradually with temperature up to 70°C. Accordingly, these films can be used without correction at temperatures between 0 and 45°C, but it is required at higher temperatures.

Pre-Irradiation Stability

The color stability before irradiation of RB/PVA film ([chloral hydrate] = 10 phr, [RB] = 0.32 phr) was tested by storing films at 35% RH in dark at room temperature and under laboratory fluorescent lights. The absorbance of the films was measured at 563 nm wavelength at different times during the pre-irradiation storage for a period of 75 days and the change in absorbance as a function of storage time relative to that before storage (zero time) is shown in Figure 12. The films show a very good stability in dark and light where ± 3% change in absorbance is obtained over all the 75-storage period.

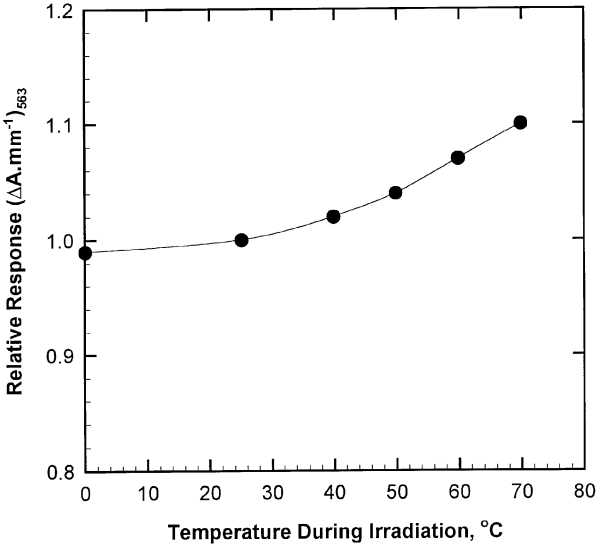

FIGURE 11 Variation of relative response of RB/PVA films ($\Delta A \cdot \mathrm{mm}^{-1}$ at 563 nm) as a function of temperature during irradiation to a dose of 6.2 kGy. [RB] = 0.32 phr; [chloral hydrate] = 10 phr.

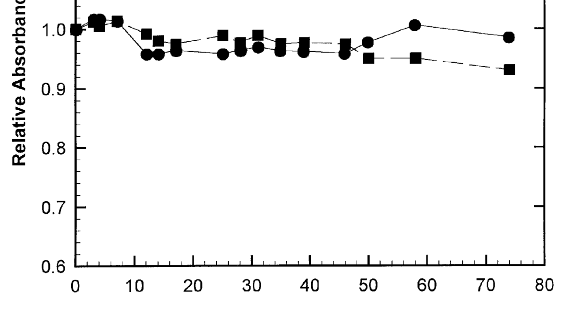

FIGURE 12 Pre-irradiation stability of RB/PVA films stored in dark and light at room temperature. [RB] = 0.32 phr; [chloral hydrate] = 10 phr; λ_{max} = 563 nm.

The unirradiated films were then read spectrophotometrically after a storage period of 12-months in dark and only a decrease in absorbance (at 563 nm wavelength) of about 5% of its initial value was noticed.

Post-Irradiation Stability

RB/PVA films ([chloral hydrate] = 10 phr, [RB] = 0.32 phr), irradiated to a dose of 6.2 kGy were stored at 33% RH in dark at room temperature and under laboratory fluorescence lights. These films were readout spectrophotometrically at 563 nm wavelength at different intervals of time during the post-irradiation storage period of 30-days, and the change of ($\Delta A \cdot \mathrm{mm}^{-1}$) as a function of storage time relative to that before storage (zero time) is shown in Figure 13. It can be seen that, the response of the film stored in dark is stable for about two weeks after irradiation and after that decreases gradually to the end of the storage period. On the other hand, the response of the stored film in laboratory fluorescent light increases gradually after irradiation to about 15% and it reaches the maximum after about 10 days storage, then begin to decrease again to the end of the storage period.

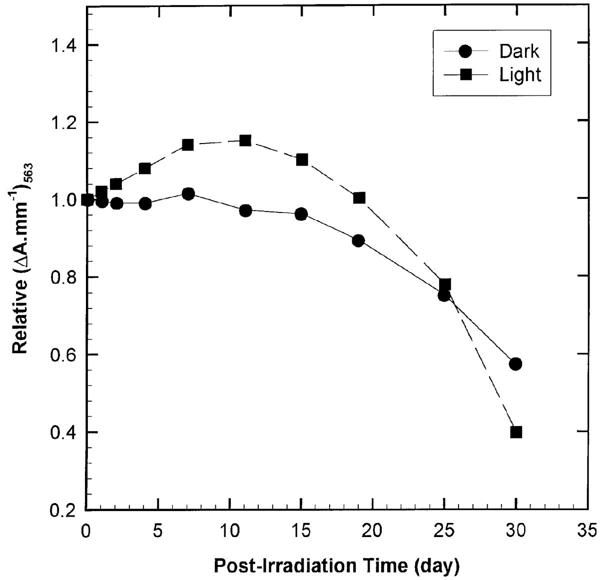

FIGURE 13 Post-irradiation stability of RB/PVA films stored in dark and light at room temperature. [RB] = 0.32 phr; [chloral hydrate] = 10 phr; $\lambda_{\max} = 563\,\mathrm{nm}$.

CONCLUSION

RB/PVA films made from PVA incorporating rose Bengal dye with and without chloral hydrate are useful as radiation dosimeters in different dose ranges depending on the concentration of chloral hydrate. Although, the response of these films depends on the relative humidity during irradiation, they are highly stable for long times before and after irradiation when stored in dark and are not affected by temperature changes during irradiation in the range from 0°C to 45°C. The properties of the prepared films suggest their useful application for routine monitoring and dose mapping in radiation processing. They are easy to prepare in laboratory and do not require toxic solvents in preparation.

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