



## EFFECT OF GAMMA RADIATION ON A MIXED DYE FILM AND ITS POSSIBLE USE AS A RADIATION DOSIMETER

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Dyed poly(vinyl alcohol) (PVA) films prepared by a simple technique of casting aqueous PVA solution incorporating a mixture of two dyes namely chlorophenol red (CPR) and Quinaldine Red (QR) on a horizontal glass plate are useful as high dose dosimeter in the dose range 2-30 kGy range. The colour of these films change from deep red to yellow when exposed to gamma radiation. Chloral hydrate was added with different concentrations. The response of the prepared films can be modified either by change of chloral hydrate concentration or ratio of the two dyes. As a result, these films can be used as a dosimeter in two dose ranges. The dosimetric parameters, e.g.; dose response, effect of relative humidity on response as well as pre- and post-irradiation stability of these films have been investigated.

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### Introduction

Ionizing radiation is commonly used for industrial applications of food preservation, blood irradiation, and medical devices sterilization. Such applications require dosimeters with higher accuracy to quantify the absorbed doses and indicators to detect visually the radiation exposure. Many dyed poly films had been developed and investigated for possibility of their being used to measure absorbed dose in nuclear reactors and doses of X rays, gamma rays and electron beam.<sup>1-6</sup> Recently a new thin plastic film dosimeter has been prepared from poly vinyl alcohol mixing with dyes. This film is considered to be as radiation dosimeter of promise.<sup>7</sup> Dyed polymer films of PVA-QR and poly(vinyl butyral) (PVB) containing QR are useful as routine high dose dosimeter. These flexible plastic film dosimeters are bleached when exposed to gamma rays. Films of PVA or PVB dyed with QR are useful dosimeter in dose range 10-90 kGy and 30-200 KGy respectively.<sup>8</sup> PVA containing CPR and chlorine containing substance (chloral hydrate) is useful for low doses application. CPR in PVA films change its color from purple to yellow by irradiation and the useful dose range extend up to 3.5 kGy.<sup>9</sup> New radiation sensitive indicators consisting of PVA film containing pH indicator dye and a water soluble chlorine containing substance have been developed.<sup>10</sup> Mixed dye film containing 2,6-dichlorophenol indophenol sodium salt and Cresol Red was investigated to be useful dosimeter in dose range up to 17 kGy and another dose range up to 50 kGy.<sup>11</sup>

The main goal of the present work is the investigation of mixed dye film of two dyes chlorophenol red (CPR) and Quinaldine Red (QR) to be a dosimeter for sterilization, also it describes the influence of humidity during irradiation as well as stability before and after irradiation on the film response.

### Experimental

#### Preparation of stock solution dyes

The stock solution of QR indicator (product of E-Merck, Molecular Weight 430.352) was prepared by dissolving 0.08 g in 50 mL 95 % ethanol. The stock solution of sodium salt of CPR indicator (products of Sigma & Aldrich, molecular weight 423.27) was prepared by dissolving 0.08 g CPR in 1.9 mL of aqueous solution of 0.1 N NaOH, then the volume was completed by 95 % ethanol in a 50 mL volumetric flask.

#### Preparation of CPR/QR /PVA mixed dye films

Films were prepared by dissolving 9 g of PVA (average M.wt 125,000, fully hydrolyzed 99-100 % product of Sigma) in 180 mL of double distilled water. Complete dissolution was attained by stirring for 3 h at 60 °C followed by continuous stirring for 2 h at room temperature and then left to cool.

The polymeric solution was divided into six parts, each of 30 mL volume to prepare six films. The first three parts contain 4 mL of CPR, 2 mL of QR from stock solution of both dye and different chloral hydrate concentrations as (0.2, 0.3, 0.4 g) respectively. The other three parts contain 2 mL CPR and 4 mL QR from stock solution of both dyes and different chloral hydrate concentrations as (0.2, 0.3, 0.4 g) respectively.

All solutions were well-stirred at room temperature for about 3 h in order to obtain a uniformly mixed solution. Each solution was poured on 15x15 horizontal glass plate and dried at room temperature for about 48 h, after stripping the films were cut into 1x1 cm pieces. Then films stored in dark at room temperature for further investigation. The thickness of the films was found to be 0.03±0.005 (σ). Six polymer films were obtained; three of them contain 0.42 phr (part per hundred) CPR and 0.21 QR phr and 13.2, 19.8, 26.4 phr chloral hydrate. The other three films contain 0.21 phr CPR and 0.42 phr QR and 13.2, 19.8, 26.4 phr of chloral hydrate.

### Irradiation sources ( $\gamma$ -irradiation)

The Gamma chamber 4000A  $^{60}\text{Co}$  irradiation facility (manufactured at Bhabha Atomic Research Center, India) is a compact and self-contained irradiation unit offering an irradiation volume of approximately  $4000\text{ cm}^3$ . The activity of this irradiation facility was  $15,000\text{ Ci}$  at the time of installation (15 June 2006). The source cage holds the radiation source pencils vertically and symmetrically along its periphery. The cage can hold a maximum of 48 pencils. Each pencil contains seven aluminium clad  $^{60}\text{Co}$  slugs.

The sample chamber is the central part of the drawer and has  $16\text{ cm}$  inside diameter and  $21\text{ cm}$  height. The central drawer is raised or lowered by means of an electric motor. The movement of the central drawer is controlled from the control panel of the irradiation chamber, which incorporates an analogue timer.

The absorbed dose rate in the irradiation facility 400A was measured by using reference alanine dosimeter and was found to be in the range  $2.25$  to  $2.1\text{ kGy h}^{-1}$  during the overall time period of the experimental part.

### UV/VIS spectrophotometer

The absorption spectra of non-irradiated and irradiated films were measured throughout the wavelength range  $200$ – $800\text{ nm}$  using UVIKON860 spectrophotometer. These films were placed into the holder designed especially for reproducible fitting into the sample beam of the instrument. The absorbance spectra were scanned to determine the wavelengths (nm) of the maximum absorbance ( $\lambda_{\text{max}}$ ) and the change in absorbance for UV/VIS spectra were recorded.

## Results and Discussion

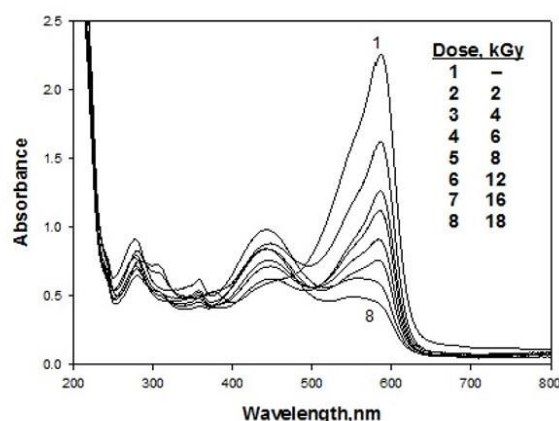
### Absorption spectra

The absorption spectra of the PVA film containing a mixture of  $0.42\text{ phr}$  of CPR and  $0.21\text{ phr}$  of QR in presence of  $26.4\text{ phr}$  chloral hydrate were recorded before and after irradiation. The spectrum of the above film is represented in Figure 1 for non-irradiated and irradiated film of different absorbed doses.

The absorption spectrum of the non-irradiated film shows a main absorption peak in the visible region peaking at  $586\text{ nm}$  characteristic of the dark red colour composed from both QR and CPR dyes. The colour of the film changes from dark red to yellow. At the beginning of irradiation gradual decrease in absorbance at  $586\text{ nm}$  takes place due to the bleaching of QR. At  $4\text{ kGy}$  orange colour appears due to the radiolysis product resulting of chloral hydrate which in turn leads to gradual transformation of CPR to its acidic form.

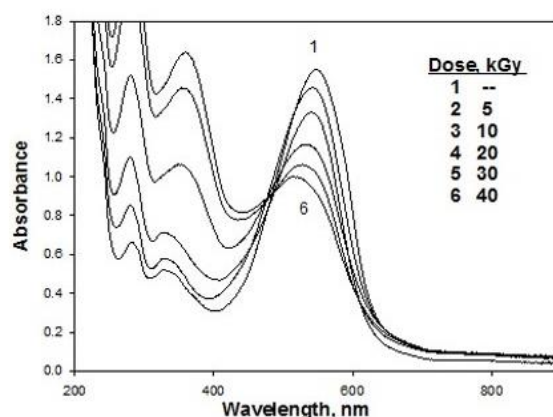
Complete irradiation of this film leads to complete bleaching of QR and complete transformation of CPR to acid form at  $18\text{ kGy}$ . It has been previously reported that complete transformation to acidic form for CPR occurs at  $3$ – $5\text{ kGy}$ . **Hiba! A könyvjelző nem létezik.** and complete bleaching of QR up to  $90\text{ kGy}$ . **Hiba! A könyvjelző nem létezik.** In the present work, the dose range of CPR was

extended to be  $18\text{ kGy}$  and that of QR to  $18\text{ kGy}$ . This behaviour is attributed to the high sensitivity of QR towards radiolysis products, so it was consumed in the degradation process and causes retardation of the transformation of CPR.



**Figure 1.** The absorption spectra of CPR/QR/PVA containing chloral hydrate for non-irradiated and irradiated films to different absorbed doses. [CPR] =  $0.42\text{ phr}$ , [QR] =  $0.21\text{ phr}$ , [Chloral hydrate] =  $26.4\text{ phr}$ .

Figure 2 shows the absorption spectra for the PVA film contains a mixture of  $0.21\text{ phr}$  CPR and  $0.42\text{ phr}$  of QR in the presence of  $26.4\text{ phr}$  chloral hydrate. The absorption spectra of non-irradiated and irradiated film show a main absorption peak at  $548\text{ nm}$ . Gradual decrease in absorbance at  $548\text{ nm}$  without any appearance of yellow color, indicating that the complete consuming of radiolysis products in bleaching reaction of QR only.



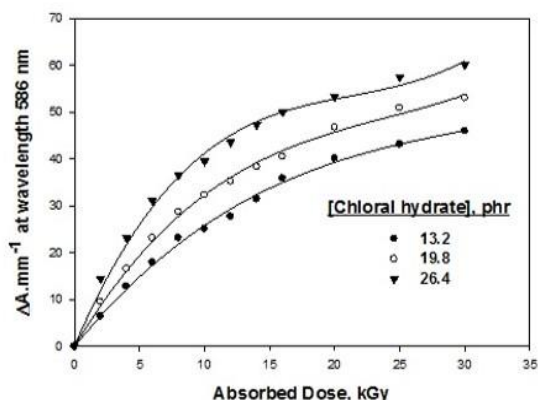
**Figure 2.** The absorption spectra of CPR/QR/PVA containing chloral hydrate for non-irradiated and irradiated films to different absorbed doses. [CPR] =  $0.21\text{ phr}$ , [QR] =  $0.42\text{ phr}$ , [Chloral hydrate] =  $26.4\text{ phr}$ .

### Response curve

Figure 3 shows the dose response curves of three films containing  $0.42\text{ phr}$  of CPR and  $0.21\text{ phr}$  of QR in presence of different chloral hydrate concentrations ( $13.2$ ,  $19.8$ ,  $26.4\text{ phr}$ ).

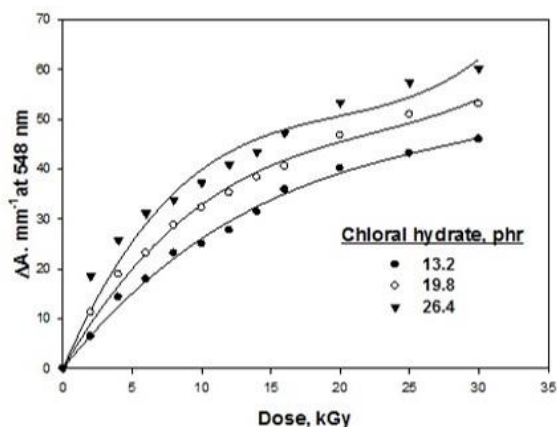
The response curves were established in terms of change in optical density measured at  $586\text{ nm}$  per unit thickness  $\Delta A\text{ mm}^{-1}$  against the absorbed dose ( $\Delta A = A_0 - A_i$ ), where  $A_0$  and  $A_i$  are values of optical absorbance at  $586\text{ nm}$  for non-irradiated and irradiated films respectively. Each point on

the response curve represents the mean average of the three measurements (three dosimeter films irradiated to the same dose). It can be noticed that all the curves show the same trend, but they differ in slope value. The slope value increases with the increase of chloral hydrate concentrations.



**Figure 3.** Change of  $\Delta A \cdot \text{mm}^{-1}$  of CPR/QR/PVA films containing different chloral hydrate concentrations. [CPR]= 0.42 phr, [QR] = 0.21 phr, at  $\lambda_{\text{max}} = 586 \text{ nm}$ .

Figure 4 shows the dose response curve of another three films containing 0.21 phr of CPR and 0.42 phr of QR and different of chloral hydrate concentrations 13.2, 19.8, 26.4 phr at wavelength 548 nm. It is noticed that the response value increases with the increases of chloral hydrate concentrations and reach saturation at different absorbed doses depending on the chloral hydrate concentrations.



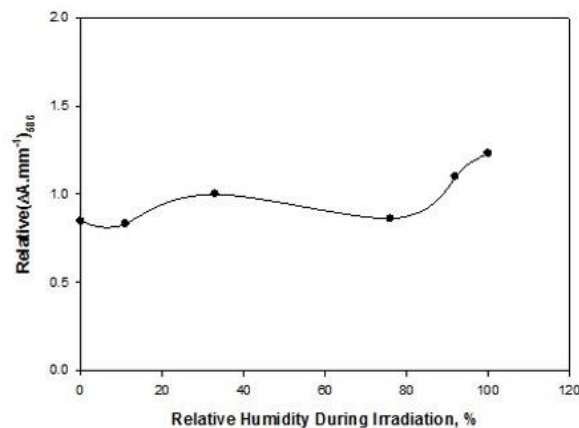
**Figure 4.** Change of  $\Delta A \cdot \text{mm}^{-1}$  of CPR/QR/PVA films containing different chloral hydrate concentrations. [CPR]= 0.21 phr, [QR] = 0.42 phr, at  $\lambda_{\text{max}} = 548 \text{ nm}$ .

From the present study, we can concluded that the preferred concentration of both dyes was 0.42 phr CPR and 0.21 phr QR, in which the degradation reaction and acidic formation takes place, the color change from red to yellow is obviously illustrated.

#### Humidity during irradiation

The effect of relative humidity (RH) during irradiation on response of CPR/ QR/PVA film, [CPR] = 0.42 phr, [QR] = 0.21 phr and [chloral hydrate] = 26.4 phr was investigated

by irradiating the films to a dose of 10 kGy at different relative humidity (0, 33, 54, 76, 92 and 100 % RH). The different relative humidity was maintained by using different saturated salt solutions.<sup>12</sup> The films were stored before irradiation for 48 h period under the same relative humidity conditions as maintained during irradiated, so that the equilibrium moisture content in dosimeter is established during irradiation.

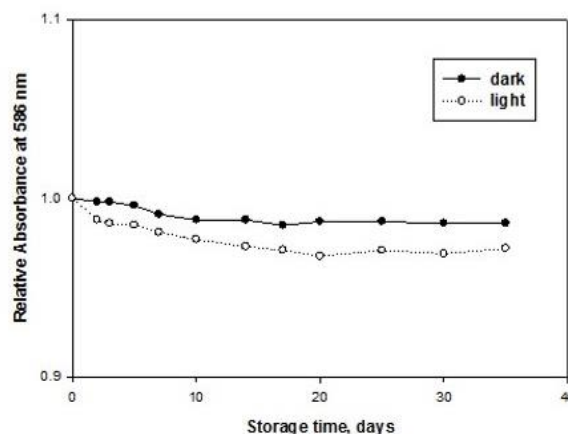


**Figure 5.** Variation in response ( $\Delta A \cdot \text{mm}^{-1}$ ) of CPR/QR/PVA films as a function of relative humidity during irradiation.  $\lambda_{\text{max}} = 586 \text{ nm}$ , dose= 10 kGy.

Figure 5 shows the variation in response ( $\Delta A \cdot \text{mm}^{-1}$ ) as a function of percentage relative humidity during irradiation relative to that at 33 %. It can be seen that there is no appreciable effect of RH on the response in the range from 0-75 % RH. At higher RH more than 75 % the response increase gradually with increase of RH up to 92 % by 1.5 % . To avoid the effects of higher humidity levels, the films should be stuffed and sealed under controlled humidity conditions in the range 0-75 % or, if that impractical, one should maintain conditions of calibration as close as possible to the conditions of use.

#### Pre-irradiation stability

The colour stability of CPR/QR/PVA film was tested before irradiation by storing the film at 35 % RH at room temperature under laboratory fluorescent light and in the dark.

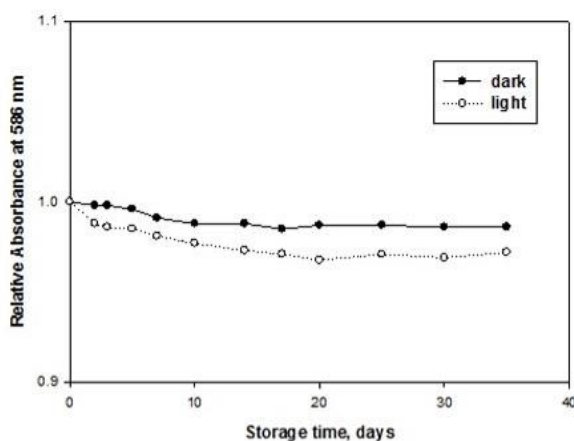


**Figure 6.** Pre-irradiation stability of CPR/QR/PVA films in terms of relative change in absorbance as a function of storage time.

Figure 6 represents the change in relative absorbance of the tested film, by measuring the absorbance at 587 nm at different time intervals during the storage period of 30 days. It can be seen that, the film exhibits excellent stability before irradiation, where the variation in the absorbance during the 30 days storage period is less than  $\pm 3.2\%$  at light and less than  $\pm 1.4\%$  at dark.

#### Post irradiation stability

Film of CPR/QR/PVA containing chloral hydrate, [chloral hydrate] = 26.4 phr irradiated to 10 kGy was stored at 35 % RH, room temperature (25 °C) under laboratory fluorescent lights and in the dark. The absorbance of the film was measured at 586 nm wavelength at different intervals time during the post- irradiation storage period of 30 days. The change in absorbance at 586 nm wavelength as a function of storage time relative to that before storage is shown in figure7. From the figure, it can be seen that, the absorbance of the film at 586 nm decrease gradually with nearly 2 % in the dark and 5 % at light during the first week then the absorbance tends to be stable up to the end of the storage period.



**Figure 7.** Post-irradiation stability of CPR/QR/PVA films in terms of relative change in absorbance as a function of storage time.

#### Conclusion

By irradiating CPR/QR/PVA films containing chloral hydrate, the colour changes from dark red to yellow indicating the acid formation. These films can be used as dosimeter in dose range 0-18 kGy. Two reactions occur in this film, first is bleaching reaction of QR and then acid formation of CPR. At 3 kGy colour change to orange due to radiolysis product resulting from chloral hydrate causes the gradual transformation of CPR to its acidic form. Then upon irradiation complete bleaching of QR and complete transformation of CPR to acid form till 18 kGy.

Addition of QR to CPR increase the dose range to 18 kGy instead of 3.5 kGy in case of CPR films and decrease the dose range to 18 kGy instead of 90 kGy in case of QR films. The best dyes ratio were 0.42 phr CPR and 0.21 phr QR, in which the degradation reaction and acidic formation takes place. The dose range depends on the chloral hydrate concentrations. The films have negligible humidity effects in range of 0-75 % RH as well as good pre and post irradiation stability. These films are easy to prepare so it is useful to use in large scale production and application for routine irradiation processes in moderate and high dose irradiation.

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