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Development of a plastic dosimeter for industrial use with high doses

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ABSTRACT

These films contain a mixture of two dyes namely bromothymol blue (BTB) and methyl orange (MO) indicator with different concentration of alanine in poly(vinyl alcohol). The color of this film changes from green to pale yellow. The response of these films can be modified by changing the alanine concentration and the ratio of the two dyes. As a result, these films can be used as a dosimeter in high dose range. The dosimetric parameter, e.g. dose response, effect of relative humidity, pre- and post-irradiation stability of these films are investigated.

INTRODUCTION

Radiation bleachable organic dyes were widely investigated (Ebraheem et al., 2005). For dose monitoring in radiation processing, the polymeric dyed flexible films are considered to be most commonly used as dosimeters, indicators (Abdel-Rehim and Abdel-Fattah, 1993) and for monitoring the absorbed dose delivered by electron beams and gamma rays (Kovaces et al., 2002). (Ueno 1988) developed a radiation dosimeter from acid indicators by coating a high molecular weight polymer support (e.g. polyester film) with a composition containing a halogen-containing polymer (e.g. PVC), a pigment which changes color with the changes of PH and basic material (e.g. KOH in EtOH). A chlorine-containing polymer is not necessary for this reaction to occur.

A similar color change can be produced in chloro-alkanes are present in the dye containing matrix (Whittaker, 1988). A system of interest in dosimetry, since it offers the possibility of a very sensitive dosimeter, is an aqueous, air-saturated, solution of chloral hydrate containing 0.001-1 M $CCl_3CH(OH)_2$. These solutions give acid products (mainly HCl) with G-values ranging from 10 to several hundreds depending upon the conditions. The high yields, concentration and dose-rate dependencies are indicative of the chain reaction initiated by radical attack upon the chloral hydrate.

For routine dose monitoring in radiation processing, the polymeric dyed flexible films are considered to be the most common ones as dosimeters, dose labels and indicators (Abdel-Rehim and Abdel-Fattah, 1993; Abdel-Rehim et al., 1985, 1991; McLaughlin et al., 1989). These dyed poly(vinyl alcohol) PVA systems are bleached by irradiation, the extent to which the color changes is used for determining the absorbed dose. Based on the idea of mixing, in poly(vinyl alcohol), two dyes having different sensitivities to radiation, and a label dosimeter system has been developed (Abdel-Rehim and Abdel-Fattah, 1993). A new radiation sensitive indicator consisting of poly(vinyl alcohol) film containing PH-indicating dye and water-soluble chlorine containing substance has been developed by Abdel-Fattah et al. (1996).

The current work deals with the investigation of a new dyed poly(vinyl alcohol) film to enable their use in high radiation processing applications.

EXPERIMENTAL

Preparation of stock solutions (BTB, MO-mix) dyes

The stock solution of the indicator was prepared by dissolving 0.04 g of both BTB and MO (product of CHMPOL, Czech Republic and RIEDEL-DEHAEN,Germany) in 25 ml distilled water.

These two stock solutions were used in the preparation process of mixed dye dosimetry film.

Preparation of (BTB, MO)/PVA mixed dye films

Fully hydrolyzed (90-100%) PVA (from Sigma), was dissolved in double distilled water at about 60^{o}C. The solution was stirred at that temperature for about 48h and after cooling, it was divided into four parts, alanine was added to three parts of polymer solution. The same amounts of BTB and MO indicators were added to four parts of the polymer solutions. All four solutions were kept well-stirred at room temperature for about 4h in order to obtain a uniformly mixed solution. Each solution was poured into a horizontal glass plate and dried at room temperature for about 48h. The different concentration of alanine as 44.66, 66.66 and 99.99 phr. After drying, the films were cut into 1 × 1 cm pieces, stored and used for different investigations. The thickness of the films was found to be 0.045±0.005 mm (1σ).

RESULTS AND DISCUSSION

Absorption Spectra

The absorption spectra for the PVA film containing a mixture of 0.533 phr of BTB and MO were recorded before and after irradiation. Fig. (1) shows the absorption spectra of unirradiated and irradiated films with different absorbed doses. The absorption spectrum of these films shows two absorption bands peaking at 622 and 405 nm. The first one is characteristic of the blue color of BTB, and the second is characteristic of the yellow color of MO indicator. The amplitude of these bands decreases gradually with the increase of dose of γ-ray photon.

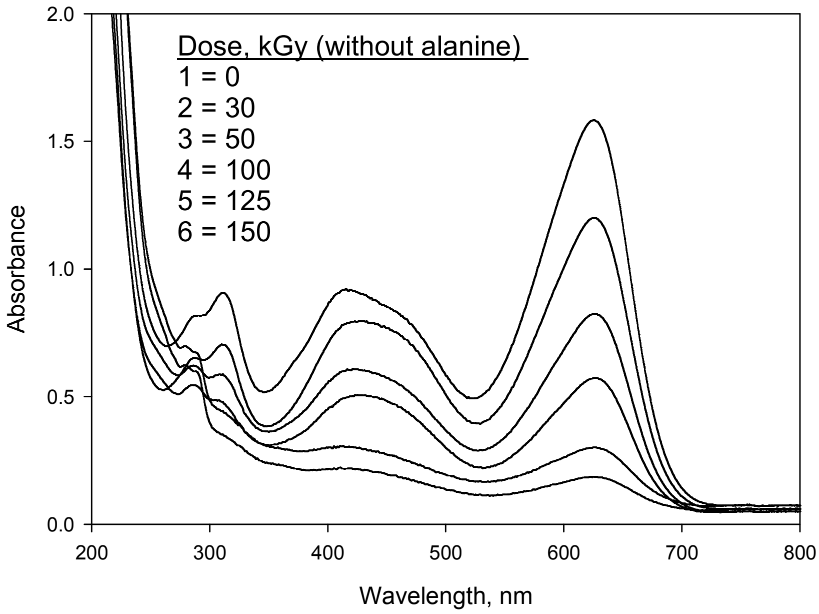

Fig. (1): The absorption spectra of (BTB-MO)/PVA films unirradiated and irradiated to different absorbed doses.

Fig. (2) Shows the absorption spectra of BTB-MO/PVA films unirradiated and irradiated to different doses. These films contain 66.66 phr alanine. Also, the amplitude of these bands decreases gradually with increase of dose of γ-ray. It was found that the useful dose range of these films 10-50 kGy. It was noticed that the bleaching reaction takes place faster within films containing alanine than that without alanine (i.e. alanine act as sensitizer).

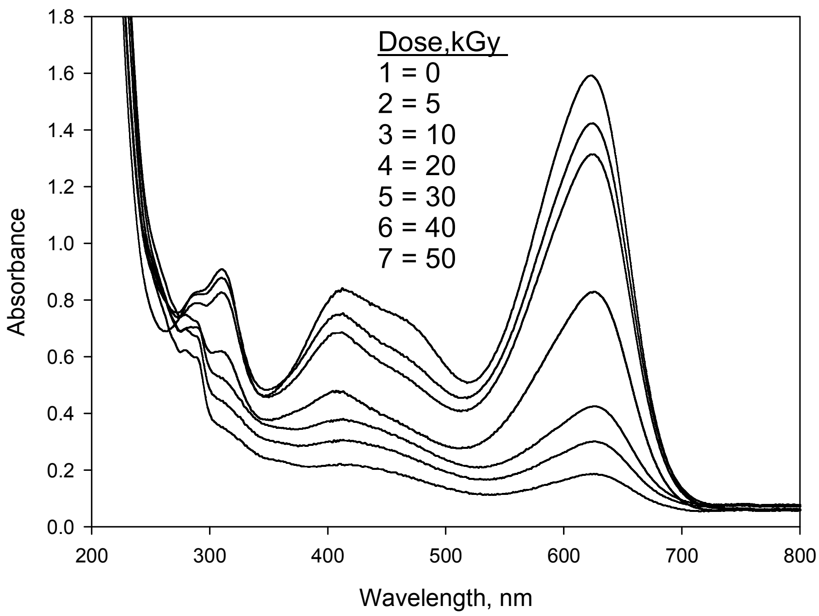

Fig. (2): The absorption spectra of (BTB-MO)/PVA films unirradiated and irradiated to different absorbed doses. [Alanine] = 66.66 phr

Response curves

Fig. (3) Shows the dose response curves of three films containing equal concentration of two combined dyes BTB and MO (0.533 phr) in the response of different concentration of alanine (44.66, 66.66 and 99.99 phr). The dose response curves were established in terms of change in optical density measured at 622 nm per unit thickness ΔA mm^{-1} against the absorbed dose ($\Delta A = A_o - A_i$), where A_o and A_i are values of optical absorbance at 622 nm for unirradiated and irradiated films. It can be noticed that all curves show the same trend, but they different in the initial slope value, which increases with increase of alanine concentration. This result reflects the sensitizing effect of alanine on radiation induced bleaching of BTB.

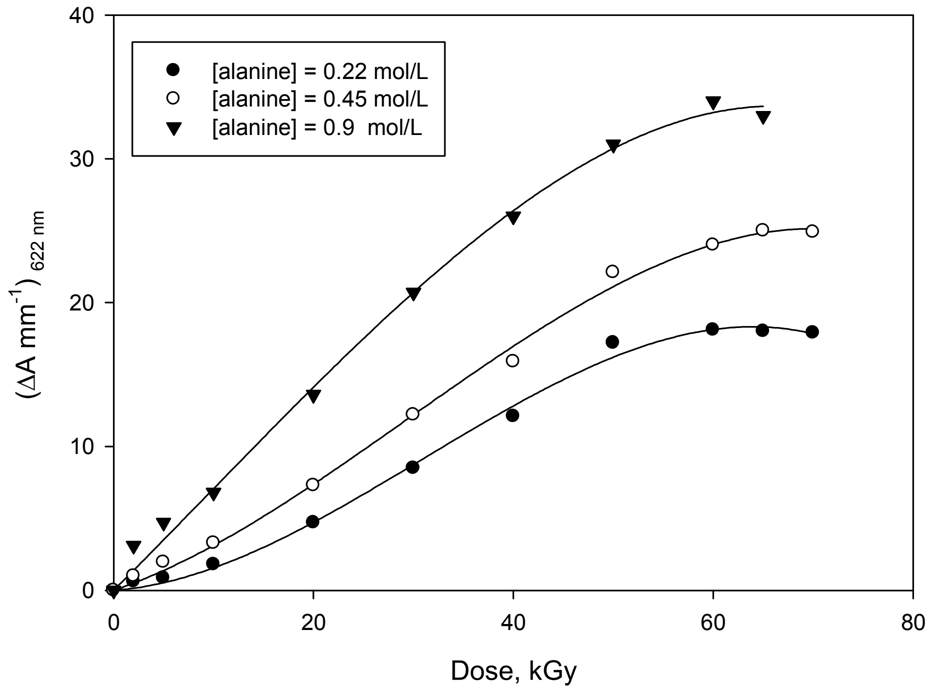

Fig. (3): Change of absorbance at 622 nm as a function of absorbed dose of (BTB-MO)/PVA films with different concentrations of alanine.

The concentration of H^+ formed in (BTB-MO)/PVA films containing different concentrations of alanine at different doses are calculated individually for each response curve. The results are plotting in Fig. (4) Between the concentration of H^+ and the absorbed dose. It can be seen from this figure that the amount of acid formed $[H^+]$ increase linearly with the increase of the absorbed dose.

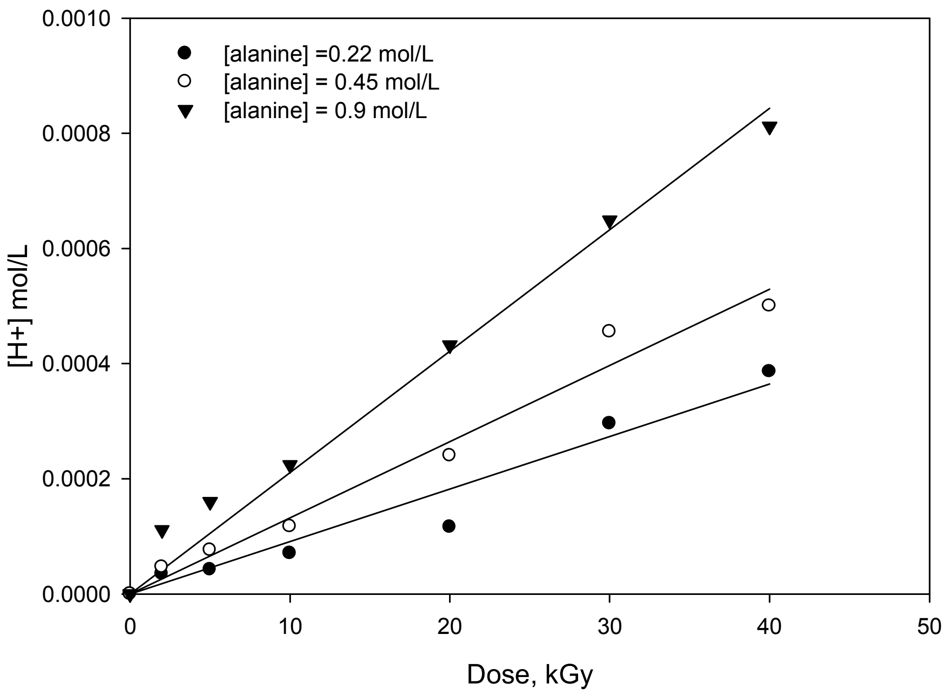

Fig. (4): Change of concentration of acid $[H^+]$ formed in (BTB-MO)/PVA films as a function of different concentrations of alanine.

Humidity during irradiation

The effect of relative humidity (RH) during irradiation on the response of (BTB-MO)/PVA films was investigated by irradiating the films 0.533 phr (BTB-MO) and 99.99 phr alanine to dose of 30 kGy at different relative humidities. The different relative humidities were maintained by using different saturated salt solutions.

The films were stored before irradiation for three days period under the same relative humidity conditions as when irradiated, so equilibrium moisture content in dosimeter is established during irradiation. Fig. (5) shows the variation in response (ΔA . mm^{-1}) at 622 nm as a function of percentage relative humidity during irradiation relative to the response value 33% relative humidity. The response is flat for relative humidities in the range of (10- 54%) with reduced sensitivity at higher humidities. It can be concluded that (BTB-MO)/PVA films can be used at negligible humidity effects on response in intermediate range of humidity from (10-54%) to avoid the effects of high humidity levels. It may also be possible to reduce the humidity influence by using sealed films under controlled intermediate humidity conditions.

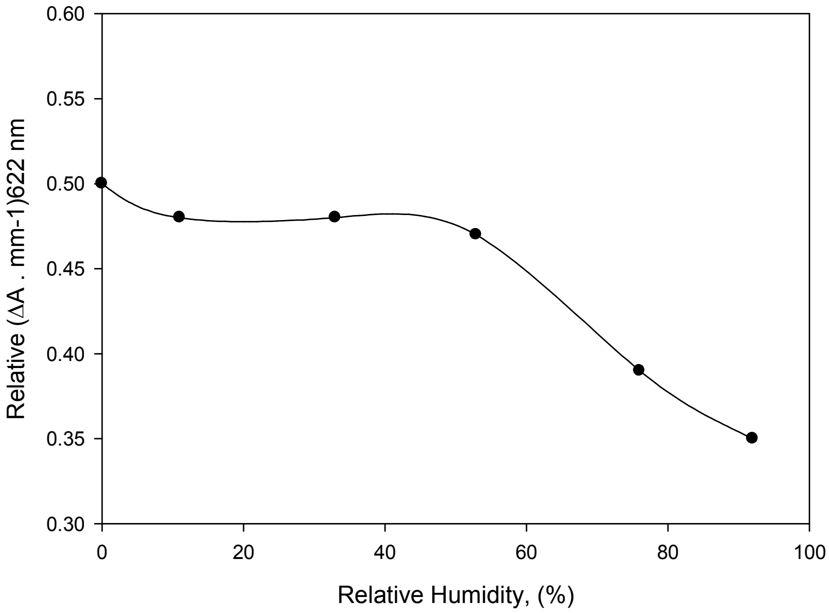

Fig. (5): Variation of response of (BTB-MO)/PVA films (at 622 nm) as a function of relative humidity during irradiation, where response in $\Delta A.\ mm^{-1}$ at 30 kGy

Post-irradiation Stability

(BTB-MO)/PVA films ([alanine] = 99.99 phr) irradiated to 30 kGy were stored immediately after irradiation, one dark and the other in indirect sunlight, both at room temperature. The films were measured spectrophotometrically at 622 nm wavelength at different intervals of time during the post-irradiation storage period of 60 days. Fig. (6) Shows the relative to the value to zero time, as a function of storage time. It can be seen that ΔA_{622} of the film stored in light decreases during the first two days after irradiation then tends to stabilize. On the other hand, the film stored in dark shows excellent stability overall the 60 days storage period.

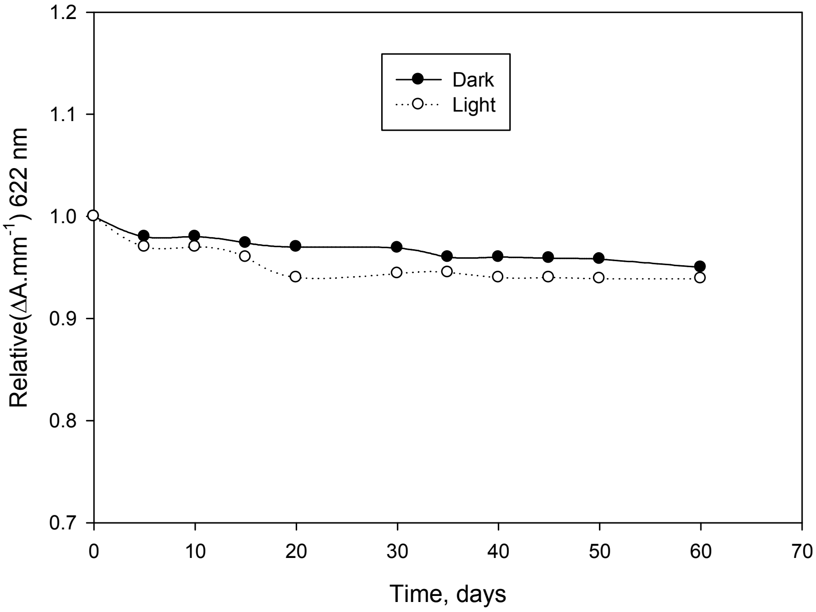

Fig.(6): Post-irradiation stability of (BTB-MO)/PVA films stored under different storage conditions

Assessment of uncertainties

To be meaningful, a measurement of gamma ray shall be accompanied by an estimate of the uncertainty in the measured value. Factors contributing to the total uncertainty may be separated into two types, type A and type B (ISO/ASTM, 2002). The first factor is associated mainly with the measuring equipment and the films and the second is mainly related to the calibration.

The reproducibility of the Unicam UV-4 spectrophotometer was determined by reading the absorbance value (at 600 nm wavelength and absorbance level 0.8) of irradiated films several times (one hundred readings per film). From the data obtained, it was found that the coefficient of variation (1 sigma\ rm) is ± 0.2%, reflecting the precision of the spectrophotometer. The reproducibility of the Minitest thickness gauge was determined by reading the thickness value for (BTB-MO)/PVA films several times (one hundred readings per film). From the data obtained, it was found that the coefficient of variation (1 sigma\ rm) is ± 0.9%. The reproducibility of the measurements of several films (10 times for each film) was found to be 0.88% (1σ).

On the other hand, the type A uncertainties (at one standared deviation, i.e. 1σ) arising during calibration over the useful response range were found to be ± 2.2% (ASTM, 1996). The combining all the components in quadrature at one standard deviation 1σ as follows:

$$U_c = \sqrt{(0.2)^2 + (0.9)^2 + (2.2)^2 + (0.88)^2}$$
$$= 2.54\%$$

The combined uncertainty (at two standard deviations, i.e. 2σ, approximately equal to a 94% confidence level) is found by multiplication of U_c (at 1σ) by two. Hence the combined uncertainty using (BTB-MO)/PVA film is 5.09%.

CONCLUSION

Films made of PVA dyed with (BTB-MO mix.) are useful radiation dosimeters in the dose range 5-150 kGy while films containing alanine are useful in the range 10-60 kGy. Although the response is discussed. The films are highly stable for long times after irradiation under different storage conditions are not affected by the humidity changes in the intermediate range of the relative humidity (10- 54%). These properties suggest them to be useful for routine monitoring and dose mapping in radiation processing. They are easy to prepare in a laboratory and do not require toxic solvents in the preparation.

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