11 *Egypt. J. Rad. Sci. Applic.*, Vol. **24**, No. 1, pp. 143-152 (2011)

# High-dose Film Dosimeter Based on a Mixture of BG and BPB Dyed Poly(vinyl butyral)

**M. El-Kelany, E. Faheem and S. Ebraheem**

*Radiation Protection and Dosimeter Dept., National Centre for Radiation Research and Technology (NCRRT), P. O. Box; 29 Nasr City, Cairo, Egypt.*

DYED poly(vinyl butyral) (PVB) film, prepared by a simple technique of casting aqueous solutions of PVB containing a mixture of brilliant green (BG), bromo phenol blue (BPB) and chloral hydrate ($CCl_3CH(OH)_2$, on a horizontal glass plate are useful as routine high-dose dosimeters. This flexible plastic film changes its colour from green to pale yellow to colourless on exposure to γ-rays photons due to the consequent lowering of pH caused by HCl generated from the radiolysis of chloral hydrate. The useful dose rang extends up to 60 kGy. Effect of different chloral hydrate concentrations on response of the film was investigated. Although the film gives excellent stability before and after irradiation when stored in the dark at room temperature beside excellent stability up to 80% relative humidity (RH). The overall combined uncertainty (at 2σ) associated with measurement of response ($\Delta A.mm^{-1}$) at 634 nm for dose range 1-60 kGy is 5.63%.

***Key words:*** Brilliant green-Bromo phenol blue), Poly(vinyl butyral), γ-rays dosimeter.

A new radiation-Sensitive indicator consisting of poly(vinyl alcohol) film containing pH-indicating dye and water soluble chlorine substance (Abdel-Fattah *et al.*, 1995) was studied. Several polymeric materials in the form of thin films have been successfully developed and used as dosimeters for routine use in gamma rays as well as electron beam radiation processing. Representative examples are radiochromic plastic films of various types (Abdel-Fattah and Miller, 1996), cellulose triacetate (Abdel-Rehim *et al.*, 1996), GafChromic dosimetry media (Chu *et al.*, 1990) and thin dyed-polymeric films (McLaughlin, 1992). Based on the idea of mixing, two dyes having different sensitivities to radiation in polymeric substrate were investigated (Abdel-Rehim and Ebraheem, 1995). 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 colour 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.

In the current work, thin films of PVB coloured with BG-BPB was investigated to be used as dosimeters for radiation processing.

## Experimental

### *Preparation of stock solution of BG-BPB*

The stock solution of mixed dye BG-BPB was prepared by dissolving 0.04 g of each (product of Aldrich) in 25 ml of absolute alcohol.

### *Preparation of the film*

7.5 grams of fully hydrolyzed PVB (99-100%) was well dissolved in 150 ml n-butanol at about 50$^{o}$C. The solution was kept well stirred for about 24h then left to cool. 1.5 ml of stock solution of mixed dye was added to the PVB solution and kept will stirred at room temperature for about 4h for uniform dye distribution. To each 30 ml of the well mixed solution, 0.3, 0.5 and 1 g of chloral hydrate (CH) were added, respectively.

The dyed PVB solution was poured onto a 15x15 cm horizontal glass plate, left to dry at room temperature for 48 h. When stripped off the film had a thickness of 0.065± 0.005mm.

### *Apparatus*

The absorption spectra of un-irradiated and irradiated films were measured using UVIKON 860 spectrophotometer. The film thickness was measured using a Digitrix-Mark II thickness gauge (precession± 1μm). Irradiation was carried out with gamma radiation in the $^{60}$Co gamma chamber 4000 Å (product of India). The absorbed dose rate in the irradiation facility was measured to be

6.384 kGy/ h, using Reference Alanine dosimeters. The electronic equilibrium condition was maintained during irradiation, through keeping the films between two polystyrene slabs of 3 mm thickness.

## Results and Discussion

### *Absorption spectra*

The absorption spectra of the BG-BPB/ PVB film with chloral hydrate were recorded before and after irradiation at different absorbed doses throughout the wavelength range 300-800 nm.

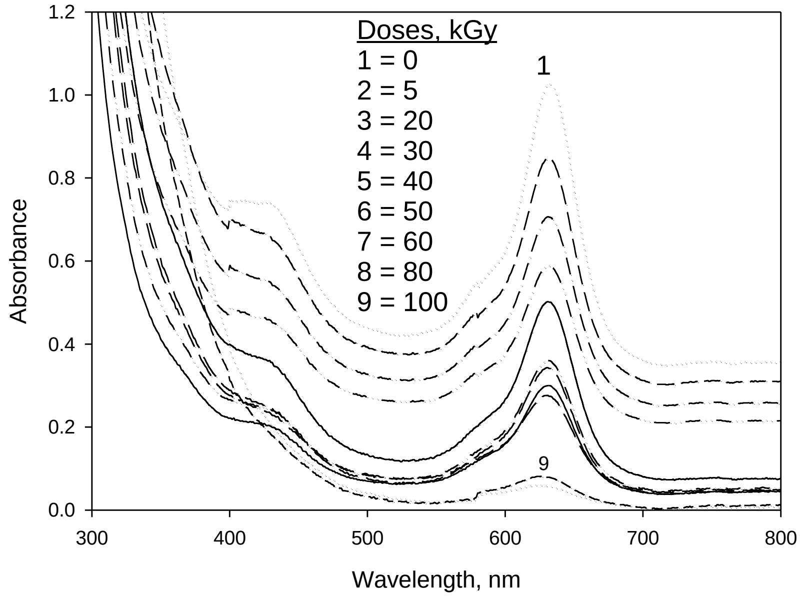


**Fig. 1. The absorption spectra of unirradiated and irradiated BG-BPB/ PVB films containing CH (66.6 phr) to different absorbed doses.**

As shown as from the above figure appearance of a shoulder at 428 nm, not addressed in films without chloral hydrate, due to interaction between green colour and yellow colour (consists of acid form resulted due to action of HCl).

The absorption spectra of un-irradiated and irradiated films show a main absorption peak at 634 nm that belongs to BG dye and a shoulder at 428 nm, that may be due to colour interfere of BG (green colour) and (yellow colour) of the acid form of BPB (not quantitative for monitoring) as shown in Fig. 1. The amplitude of the absorption band decreases gradually with increase of dose. By increasing the absorbed doses, the amplitude of the peak at 634 nm decrease till completely colour fading at about 100 kGy. From previous studies (Abdel-Fattah *et al.*, 1996) the BPB with the same concentrations used in our work indicated limited response range (maximum 5 kGy), but in our studies addition of BG extend the response to 60 kGy. Accordingly, other types of radiation processing applications could be covered such as pasteurization and sterilization of medical devices, spices and herbs irradiation ….etc. So, the BG may be behaving as dissociated material for absorbed dose leads to extend the response range to 60 kGy.

***Response curves***

Fig. 2. shows the response curves of BPB-BG/PVB films containing different concentrations of chloral hydrate (20, 33.3 and 66.6 phr). It can be noticed that the three curves have S-shape, characteristic of pH indicator in an acid base titration. Each curve reaches saturation at different dose depending on the concentration of CH.

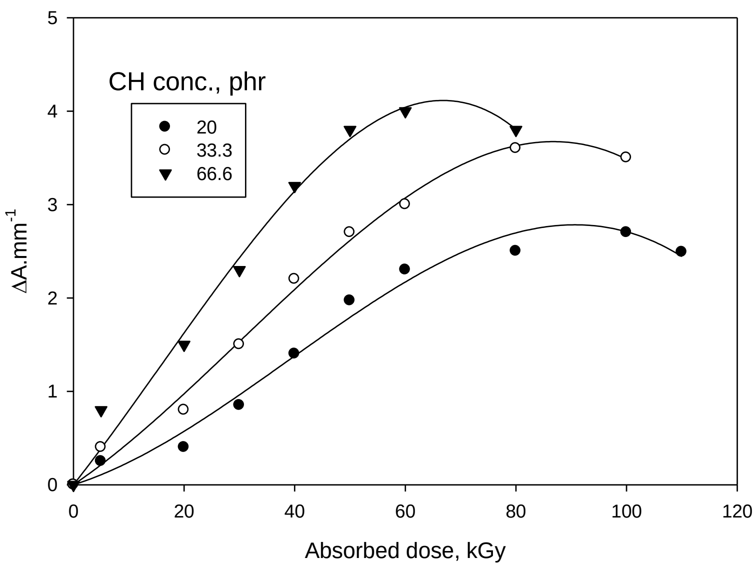


**Fig. 2. Change of ΔA.mm-1 as a function of absorbed dose of BG-BPB/ PVB films with different concentrations of chloral hydrate (at 634 nm).**

Fig. 3. shows the relation between the dose at saturation and the concentration of CH. It can be seen that the dose at saturation decreases with increase of CH from 20 up to 66.6 phr and a straight line was obtained (i.e. can be expressed as follows: D = 105.2 - 0.813 [CH]) where; D is the absorbed dose at saturation in kGy, [CH] is the concentration of chloral hydrate in phr.

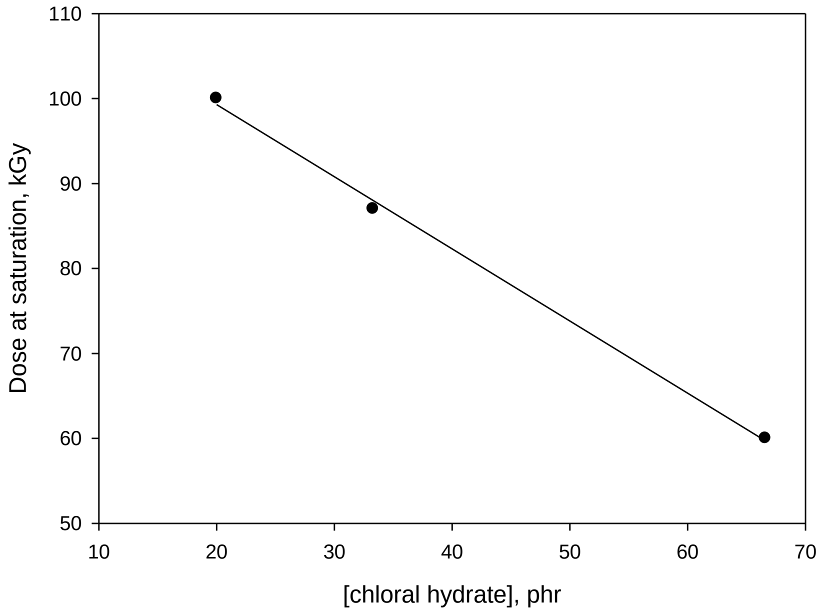


**Fig. 3. Variation of dose at saturation, kGy, of BG-BPB/PVB films as a function of concentration of chloral hydrate.**

### *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: mole/J). The G-value is calculated from the general relation (McLaughlin *et al.*, 1989).

$$G\,(Dye) = \Delta A / D.\varepsilon.\rho.b \qquad (mol/J)$$

Where, $\Delta A$ is the change in absorbance at $\lambda_{max}$, b is the optical path length (cm), $\varepsilon$ is the molar extinction coefficient at $\lambda_{max}$ (L $mol^{-1}$ $cm^{-1}$), $\rho$ is the density of dosimeter ($g.cm^{-3}$) and D is the absorbed dose (Gy).

Using the density of PVB (1.1 $g.cm^{-3}$), by using the initial curves slope in $Gy^{-1}.cm^{-1}$, the G-value at different concentrations of chloral hydrate at 634 nm are given in Fig. 4. It can be seen that the yield, increases with increase of concentration of chloral hydrate.

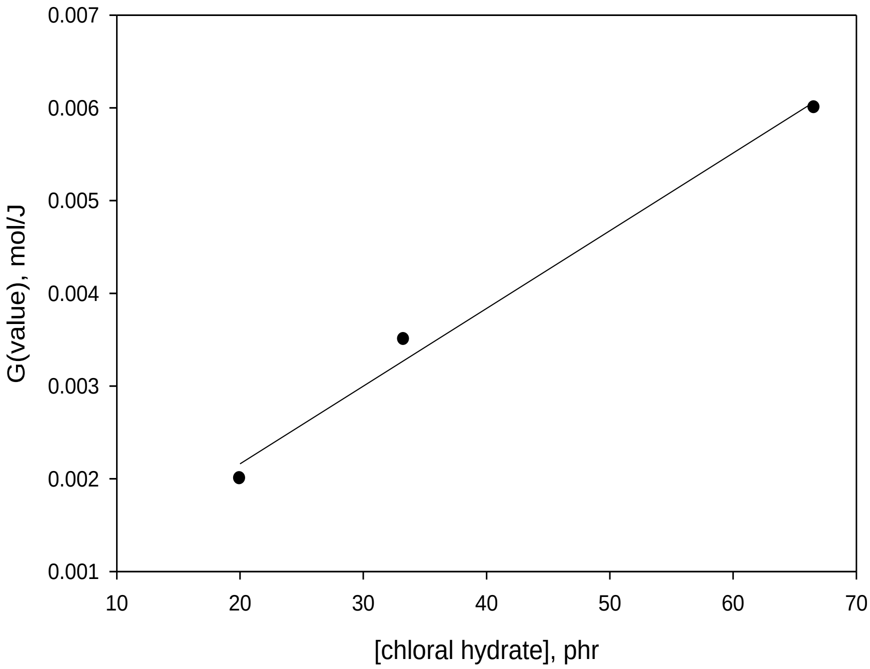


**Fig. 4. The radiation chemical yield, G(acid) versus the concentration of chloral hydrate in BG-BPB/ PVB films.**

***Humidity during irradiation***

The effect of relative-RH during irradiation on the response was investigated by irradiating BG-BPB/PVB films (40 kGy) at different relative humilities (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, except for the 0 % RH which was suspended over dried silica gel.

Fig. 5. shows the variation in response ($\Delta$A. mm$^{-1}$) as a function of percentage RH during irradiation relative to that at 33%. It was found that, for these films there is no appreciable effect in the range of relative- RH (10-80%), although the response show somewhat different sensitivity at both higher and lower humilities.

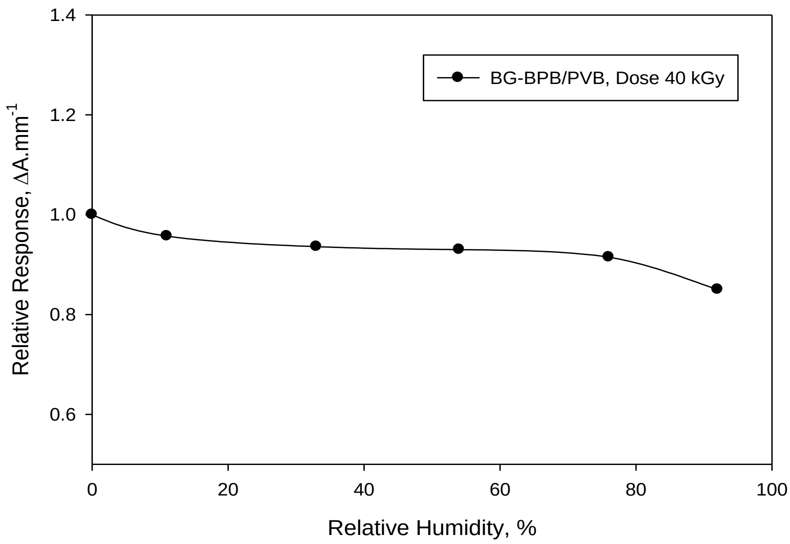


**Fig. 5. Variation of relative response of BG-BPB/ PVB films (at 634 nm) as a function of relative-RH during irradiation.**

***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 630 nm wavelength and absorbance level 1.1) of irradiated films several times (one hundred readings per film). From the data obtained, it was found that the coefficient of variation (1σ) is± 0.26%, reflecting the precision of the spectrophotometer. The reproducibility of the Digitrix MarkII thickness gauge was determined by reading the thickness value for BG-BPB/ PVB film several times. From the data obtained, it was found that the coefficient of variation (1σ) is± 0.7%. The reproducibility of the measurements of several films (10 times for film) was found to be 0.78% (1σ). On the other hand, the type A uncertainties (at one standard deviation, i.e. 1σ) arising during calibration of film over the useful response range was found to be ± 2.6%. The combined uncertainty ($U_c$) is calculated by combining all the components in quadrature at one standard deviation (1σ) as follows:

$$U_c = \sqrt{(0.26)^2 + (0.7)^2 + (2.6)^2 + (0.78)} = 2.82\%$$

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 BG-BPB/ PVB film is 5.63%.

### *Shelf-life and post-irradiation stability*

Stability measurements for the films in the dark at 25°C before irradiation were investigated during 2 month's period of time (Fig. 6). It can be seen that excellent stability for films was recorded before irradiation, but we have to left films 10 days for stability requirements. The response of irradiated films decreases gradually for about two weeks, after that it gives excellent stability in dark to the end of 60 days storage period.

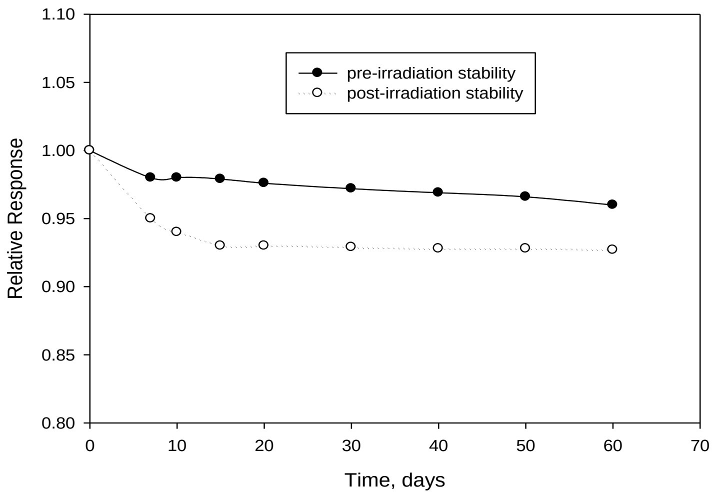


**Fig. 6. Stability before and after irradiation of BG-BPB/ PVB) films stored in the dark (at 25 °C).**

### *Conclusion*

Films made of PVB dyed with mixed dyes BG-BPB are useful radiation dosimeters in the dose range 5-60 kGy. The films are highly stable for long times in dark and are not affected by RH changes in the intermediate range of relative RH (20-60%). These films are easy to make and thus, amenable for large-scale production and application for routine irradiation processes of food and medical devices. The combined uncertainty at 2σ using BG-BPB/ PVB films was found to be 5.63%.

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*(Received: 21/08/2011;*

*accepted: 28/09/2011)*

**دراسة امكانية استعمال أفلام البولى فينيل بيوتيرال المصبوغة بخليط من البرلينت الأخضر وبروموفينول الأزرق كمقياس للجرعات الاشعاعية العالية**

**مشيرة عبد القادر الكيلانى و عصام فهيم و سيف الدين ابراهيم**

*قسم الوقاية و الجرعات الاشعاعية ، المركز القومى لبحوث و تكنولوجيا الاشعاع ، ص.ب. ٢٩ مدينة نصر ، القاهرة ، مصر.*

هذة الأفلام المتبلمرة المحضرة من عديد الفينيل بيوتيرال والذى يحتوى على خليط من صبغتى البرلينت الخضراء والبروموفينول الزرقاء و مادة الكلورال هيدرات والتى يتم تحضيرها عن طريق الصب على لوح زجاجى أفقى حيث تعتبر من الأفلام المفيدة فى قياس الجرعات الاشعاعية العالية وقد تبين أن الأفلام البلاستيكية المرنة تفقد لونها الأخضر تدريجيا أثناء تعرضها لجرعات متزايدة من أشعة جاما فى المدى أكبر من ١٠٠ كيلو جراى وذلك نتيجة للانخفاض المتتالى فى الأس الهيدروجينى بسبب تكوين نواتج حمضية ثبت انها تتكون من تشعيع المادة المحتوية على الكلور وتم تقدير الناتج الكيميائى الاشعاعى لهذة الأفلام المحضرة وأيضا دراسة حساسية استجابة هذة الأفلام لأشعة جاما وارتباطها بمدى تركيز مادة الكلورال هيدرات . كما تم دراسة تأثير الرطوبة على حساسية الأفلام الى المدى ٨٠% أثناء التشعيع.