

# Color Changes in Gamma Irradiated CR-39 and Makrofol Nuclear Track Detectors

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The effect of gamma irradiation on the color changes of CR-39 diglycol carbonate and Makrofol polycarbonate solid state nuclear track detectors was investigated. Samples from CR-39 and Makrofol polycarbonates were irradiated with gamma doses at levels between 100 and 2000 Gy. The transmission of these samples in the wavelength range 200–2000 nm, as well as any color changes, was studied. The Commission International de E'Claire (CIE units  $x$ ,  $y$  and  $z$ ) methodology was used in this work for the description of colored samples. In addition, the color differences between the non-irradiated sample and those irradiated with different gamma doses were calculated. The results indicate that both CR-39 and Makrofol detectors acquire color changes under gamma irradiation, but the Makrofol detector has more response to color change than that of CR-39.

**Keywords:** gamma irradiation, color changes, CR-39, Makrofol

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## Introduction

Demand for polymers having enhanced properties is continuously on the rise due to their use for various scientific and technological applications.<sup>1,2</sup> Irradiation of polymers has established itself as one of the most acceptable approach to alter polymer properties significantly.<sup>3–10</sup> Irradiation of polymers destroys<sup>11</sup> the initial structure by way of cross linking, free radical formation, irreversible bond cleavages etc. that results in the fragmentation of molecules and formation of saturated and unsaturated groups.<sup>8</sup> All of these processes introduce defects inside the material that are responsible for change in the optical and structural properties of the polymer.<sup>3</sup>

On the other hand, color is the attribute of reflected and transmitted light that, while based on certain arbitrarily agreed upon mathematical notation, is nevertheless scientifically derived from a systematic description of the spectral response of the human eye. The mathematical description of color, or color notation to be more precise, is based on dividing the spectral response of the human eye into three primary colors Red, Green and Blue (RGB). The RGB color space is used in color mixing, color specification and color difference measurements of colored plastics.

The study of color changes in irradiated polymers is an important technique that has been used to assess physical changes in polymers.<sup>12</sup> Factor et al,<sup>13</sup> studied the color changes in gamma irradiated polycarbonate. They reported that polycarbonate turns to moderately intense yellow color due to gamma irradiation, while when it is irradiated with electron beam, it acquires a green color. In our previous work<sup>14</sup> we used the Commission International de E'Claire methodology to study the electron beam induced color changes in cellulose triacetate. Also, we studied the effect of alpha particle irradiation on the color changes of Makrofol polycarbonate.<sup>15</sup> The Makrofol samples showed significant darkness sensitivity towards alpha particle irradiation. The aim of the present study is not only to obtain information concerning the interaction of gamma rays with both CR-39 and Makrofol nuclear track detectors but also to introduce the basis that can be used in constructing simple sensor for gamma irradiation.

## Experimental Samples

CR-39 (diglycol carbonate) sheets manufactured by Pershore, Ltd., England are used in this study. It is of density 1.32 g/cm<sup>3</sup> and thickness 500 μm. While, Makrofol DE 6–2 is a bisphenol-A polycarbonate with a chemical composition of C<sub>16</sub>H<sub>14</sub>O<sub>3</sub>. It is manufactured by Farbenfabriken Bayer A.G., Leverkusen (Germany), with an average thickness of 300 μm and density 1.23 g/cm<sup>3</sup>.

## Irradiation facilities

A <sup>60</sup>Co source (manufactured by Atomic Energy of Russia) providing a dose rate of 5.28055 kGy/h was used. The measurements were carried out 24 h after the irradiation.

## Experimental apparatus

### Color measurement

The transmission measurements were carried out using a Shimadzu UV–Vis–Nir scanning spectrophotometer, type 3101 PC. This unit measures in the wavelength range from 200 to 3000 nm. The Commission International de E'Claire (CIE units *x*, *y* and *z*) methodology was used in this work for the description of colored samples.

### Determination and calculation of the tristimulus values

The vision scientists created a special set of mathematical lights, *X*, *Y* and *Z*, to replace actual red, green and blue lights. The color matching functions for the *X*, *Y* and *Z* lights are all positive numbers and are labeled  $\bar{x}$ ,  $\bar{y}$  and  $\bar{z}$ . Every color can be matched using the appropriate amount of *X*, *Y* and *Z* light. The amount of *X*, *Y* and *Z* light needed to match a color are called the color's tristimulus values.

The CIE tristimulus values for a transmitting sample are calculated by adding the product of the spectral power distribution of illuminant, the transmittance factor of the sample and the color matching functions of the observer at each wavelength of the visible spectrum, as shown in the following equations:

$$X = k \sum P(\lambda) \bar{x}(\lambda) T(\lambda),$$

$$Y = k \sum P(\lambda) \bar{y}(\lambda) T(\lambda),$$

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$$Z = k \sum P(\lambda) \bar{z}(\lambda) T(\lambda),$$

$$k = \frac{100}{\sum P(\lambda) \bar{y}(\lambda)}$$

where  $P(\lambda)$  is the value of the spectral power distribution of the illuminant at the wavelength  $\lambda$ .  $T(\lambda)$  is the transmittance factor of the sample at the wavelength  $\lambda$  and  $\bar{x}(\lambda)$ ,  $\bar{y}(\lambda)$  and  $\bar{z}(\lambda)$  are the CIE color matching functions for the standard observer at the wavelength  $\lambda$ . The factor  $k$  normalizes the tristimulus value so that  $Y$  will have a value of 100 for a perfect white diffuser.

The 1976 CIE  $L^* a^* b^*$  (CIELAB) color space

A weakness of the CIE  $X, Y$  and  $Z$  color space is its lack of visual uniformity. Creating a uniform color space would have two major advantages. It would allow plots showing the perceptually relative positions of two or more colors in color space, and it would facilitate the creation of a good color difference ruler between two samples.

The 1976 CIE  $L^* a^* b^*$  (CIELAB) color space is widely used in the paint, plastic and textile industries, while the 1976 CIE  $L^* u^* v^*$  (CIELUV) color space is widely used in the television and video display industries.

$L^*$  correlates with perceived lightness in CIELAB color space. A perfect white would have an  $L^*$  of 100, and a perfect black would have an  $L^*$  of 0. The coordinates  $a^*$  and  $b^*$  have their history in the opponent color theory. It was proposed that three pairs of opposing color sensations produce all colors: red and green; yellow and blue; and black and white. The CIELAB coordinate  $a^*$  correlates with red ( $+a^*$ ) and green ( $-a^*$ ), while the coordinate  $b^*$  correlates with yellow ( $+b^*$ ) and blue ( $-b^*$ ). The CIELAB  $L^*$ ,  $a^*$  and  $b^*$  coordinates are calculated from the tristimulus values according to the following equations

$$L^* = 116f(Y/Y_n) - 16,$$

$$a^* = 500[f(X/X_n) - f(Y/Y_n)],$$

$$b^* = 200[f(Y/Y_n) - f(Z/Z_n)].$$

In which  $X, Y$  and  $Z$  are the tristimulus values and the subscript  $n$  refers to the tristimulus values of the perfect diffuser for the given illuminant and standard observer;  $f(X/X_n) = (X/X_n)^{1/3}$  for values of  $(X/X_n)$  greater than 0.008856 and  $f(X/X_n) = 7.787(X/X_n) + 16/116$  for values of  $(X/X_n)$  equal to or less than 0.008856; and the same with  $Y$  and  $Z$  replacing  $X$  in turn.

The CIELAB color difference,  $\Delta E$  is given by<sup>16,17</sup>

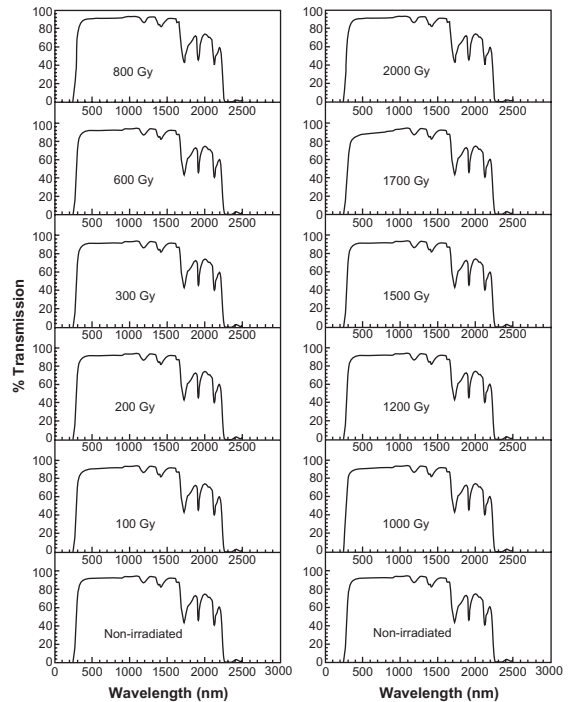
$$\Delta E = [(L^*_1 - L^*_2)^2 + (a^*_1 - a^*_2)^2 + (b^*_1 - b^*_2)^2]^{1/2}$$

The subscripts 1 and 2 refer to the irradiated and non irradiated samples.

## Results and Discussion

### Color changes in gamma irradiated CR-39 nuclear track detector

Samples from CR-39 detector were irradiated with gamma doses at levels between 100 and 2000 Gy. The % transmission spectra of these samples in the wavelength range 200–2000 nm have been investigated. The spectra appeared, for all CR-39 samples, as a band with different intensities as shown in Figure 1. The position of the peaks is almost independent on



**Figure 1.** The % transmission spectra for the non-irradiated and gamma irradiated CR-39 samples.

the gamma dose but the intensity of the bands varied with the gamma dose. Using the transmission data, both the tristimulus values and chromaticity coordinates were calculated. Figure 2 shows the variation of tristimulus values ( $X$ ,  $Y$ ,  $Z$ ) with the gamma dose. From the figure it is clear that  $X$ ,  $Y$  and  $Z$  exhibited the same trend, where they show a decrease up to a minimum value around the 600 Gy irradiated sample followed by an increase on increasing the gamma dose up to a maximum value around the 1200 Gy irradiated sample. Above 1200 and up to 2000 Gy it decreases again. Figure 3 shows the variation of chromaticity coordinates ( $x$ ,  $y$ ,  $z$ ) with the gamma dose. From the figure it is clear that  $x$  and  $y$  exhibited the same trend, where they increased with increasing the gamma dose up to 2000 Gy. The chromaticity coordinate  $z$  exhibited the opposite trend with the gamma dose.

The variation of color intercepts ( $L^*$ ,  $a^*$ , and  $b^*$ ) with the gamma dose is shown in Figure 4. The accuracy in measuring  $L^*$  is  $\pm 0.05$  and  $\pm 0.01$  for  $a^*$  and  $b^*$ . It can be seen that the color parameters  $b^*$  and  $L^*$  were significantly changed after exposure to gamma irradiation. The blue ( $-b^*$ ) color component

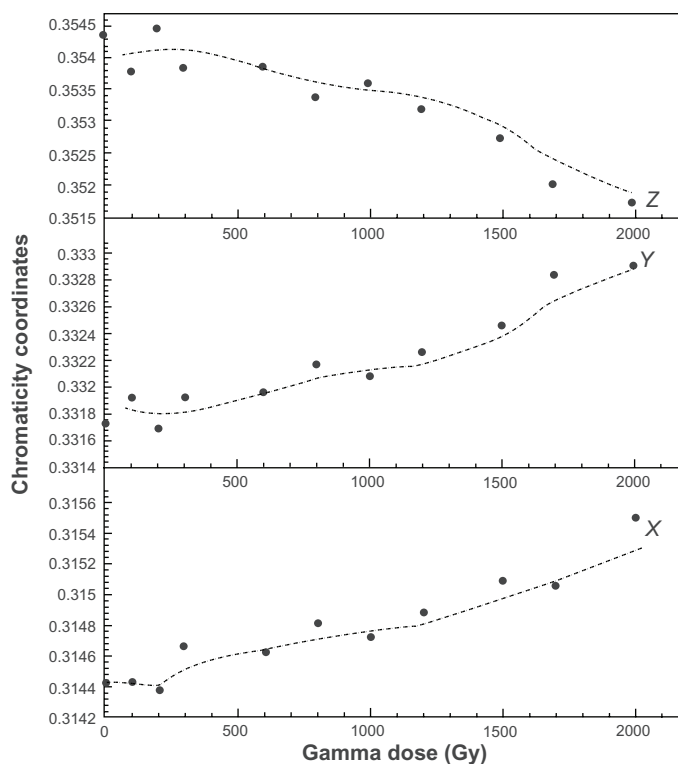


Figure 3. The variation of chromaticity coordinates ( $x$ ,  $y$ ,  $z$ ) for CR-39 detector with the gamma dose.

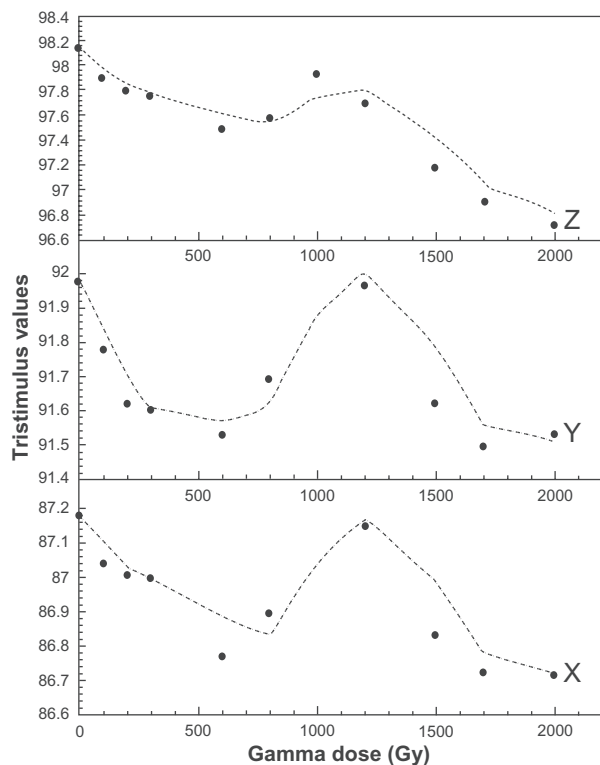


Figure 2. The variation of tristimulus values ( $X$ ,  $Y$ ,  $Z$ ) for CR-39 detector with the gamma dose.

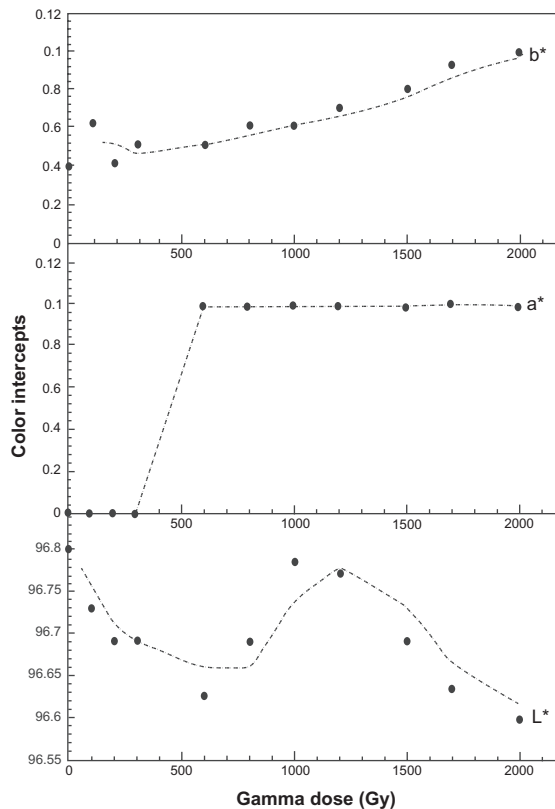


Figure 4. The variation of color intercepts ( $L^*$ ,  $a^*$ , and  $b^*$ ) for CR-39 detector with the gamma dose.

of the non irradiated film was changed to yellow ( $+b^*$ ) after exposure to gamma up to 2000 Gy. This is accompanied by a net increase in the darkness of the samples ( $-L^*$ ). At the same time, the red ( $+a^*$ ) color of the non irradiated sample was not affected by the gamma doses. Figure 5 shows the variation of the color intensity  $\Delta E$  (color difference between the non irradiated sample and those irradiated with different gamma doses) with the gamma dose. From the figure it is seen that  $\Delta E$  was greatly increased with increasing the dose, and accompanied by a significant increase in the yellow color components ( $+b^*$ ). This indicates that the CR-39 polymer has a response to color change by gamma irradiation. These changes in color can be attributed to the trapping of the excited free radicals that are formed by ionization.<sup>18</sup> Also, the trapped free radicals resulting from radiation-induced rupture of polymer molecules have electrons with unpaired spin. Such species may also give optical coloration.

### Color changes in gamma irradiated Makrofol nuclear track detector

Samples from Makrofol detector were irradiated with gamma rays in the dose range 100–2000 Gy. The % transmission spectra of these samples in the wavelength range 200–2000 nm have been investigated. The spectra appeared, for all Makrofol samples, as a band with different intensities as shown in Figure 6. Figure 7 shows the variation of tristimulus values ( $X$ ,  $Y$ ,  $Z$ ) with the gamma dose. From the figure it is clear that  $X$ ,  $Y$  and  $Z$  exhibited the same trend, where they show a non monotonic trend on gamma irradiation up to 600 Gy, then increases up to a maximum value around the 1300 Gy irradiated sample. Above 1300 and up to 2000 Gy it decreases.

Figure 8 shows the variation of chromaticity coordinates ( $x$ ,  $y$ ,  $z$ ) for Makrofol with the gamma dose. From the figure it is clear that  $x$  and  $y$  exhibited the same trend, where they show a decrease up a maximum value around 600 Gy irradiated sample, followed by an increase on increasing the gamma dose up to 1200 Gy. Above 1200 and up to 2000 Gy, it decreases again. The chromaticity coordinate  $z$  exhibited an opposite trend.

The variation of color intercepts ( $L^*$ ,  $a^*$ , and  $b^*$ ) of Makrofol detector with the gamma dose is shown in Figure 9. It can be seen that the color parameters

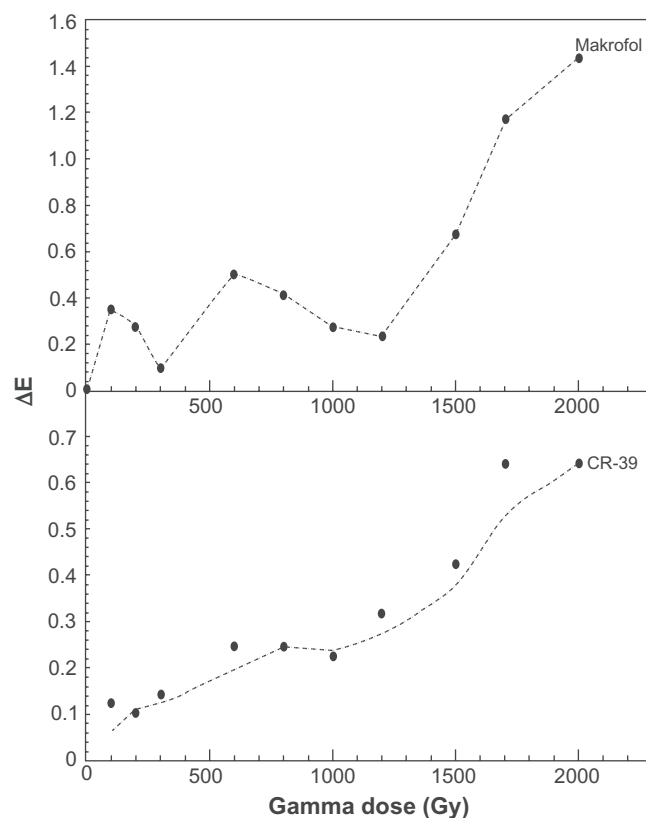


Figure 5. The variation of the color intensity  $\Delta E$  for both CR-39 and Makrofol detectors with the gamma dose.

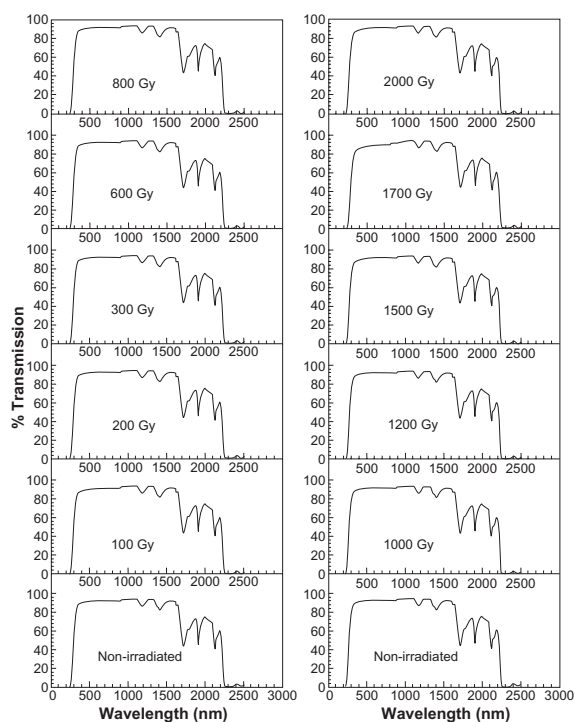
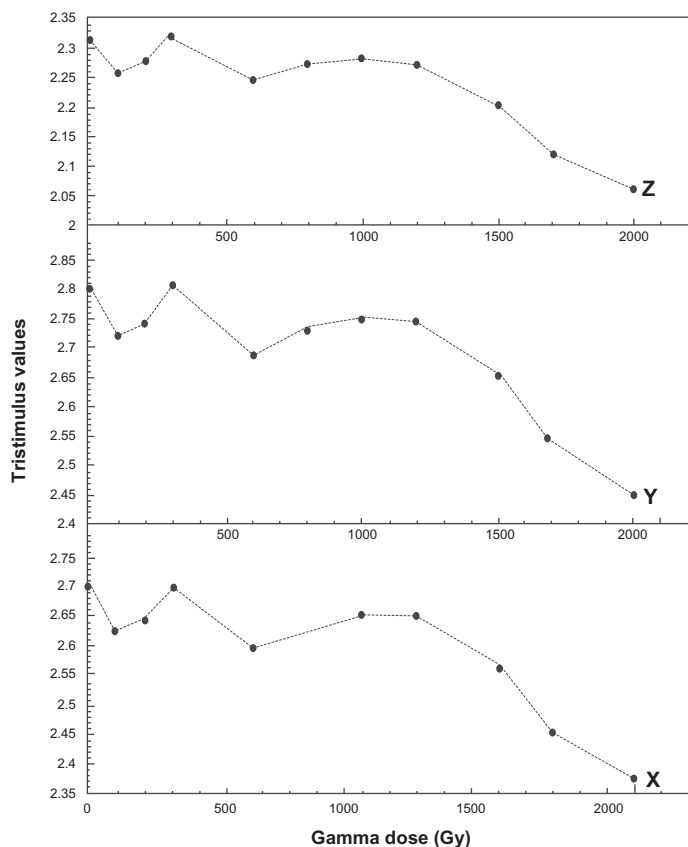
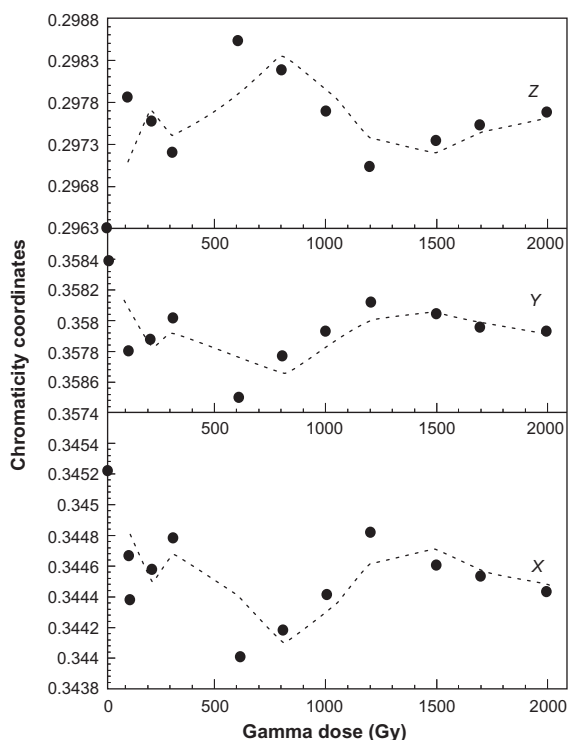


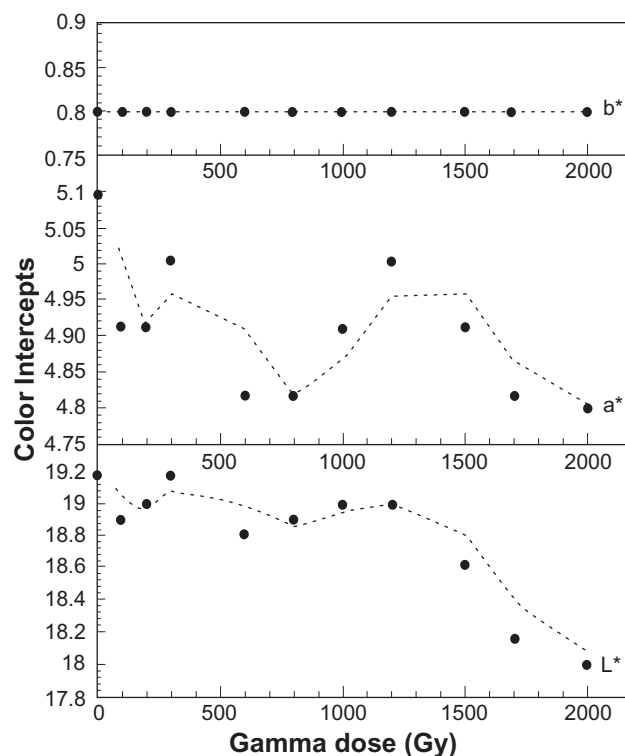
Figure 6. The % transmission spectra for the non-irradiated and gamma irradiated Makrofol samples.



**Figure 7.** The variation of tristimulus values (X, Y, Z) for Makrofol detector with the gamma dose.



**Figure 8.** The variation of chromaticity coordinates (x, y, z) for Makrofol detector with the gamma dose.



**Figure 9.** The variation of color intercepts ( $L^*$ ,  $a^*$ , and  $b^*$ ) for Makrofol detector with the gamma dose.

$L^*$  and  $a^*$  were significantly changed after exposure to gamma irradiation. There is a net increase in the darkness of the samples ( $-L^*$ ) and green components ( $-a^*$ ). The blue ( $-b^*$ ) color component of the non irradiated sample was not affected by the gamma irradiation. The variation of the color intensity  $\Delta E$  with the gamma dose is shown in Figure 5. From the figure it is seen that  $\Delta E$  was greatly increased with increasing the dose, and accompanied by a net increase in the darkness of the samples ( $-L^*$ ) and green components ( $-a^*$ ). The difference in color parameters between CR-39 and Makrofol detectors can be attributed to the fact that the interaction of radiation with matter depends basically on the type of the target detector (category of the absorber medium).

## Conclusion

Both CR-39 and Makrofol detectors acquire color under gamma irradiation but the Makrofol detector has more response to color changes than CR-39 detector. This means that the range of doses in which discoloration becomes noticeable varies widely depending on the chemical structure of the polymer.

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The non irradiated CR-39 and Makrofol polymers are nearly colorless; however, they showed significant color sensitivity towards gamma irradiation. The sensitivity in color change towards gamma irradiation appeared clearly in the change in the blue color component of the non irradiated CR-39 film to yellow after exposure to gamma up to 2000 Gy. This is accompanied by a net increase in the darkness of the samples. At the same time, the red color components of Makrofol detector was changed to green, accompanied by an increase in the darkness.

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