BJRS

BRAZILIAN JOURNAL
OF
RADIATION SCIENCES
09-01A (2021) 01-11

Evaluation of gamma irradition process for induction of color change in gel composite

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ABSTRACT

The gamma irradiation process of industrial and medical materials is based on radio-induced changes for specific purposes, such as sterilization or induction of color formation. In gem irradiation, for example, color changes according to the deposited dose. In this work we present results obtained by the irradiation of clinical gel/copper sulfate composite to observe the color change for further evaluation of its use as an indicator of volume dose distribution. The irradiations were performed with a Co-60 source at doses between 10-100 kGy in Gamma Irradiation Laboratory (LIG) of Centro de Desenvolvimento da Tecnologia Nuclear (CDTN). Samples were prepared in three different compositions in relation to the amount of copper sulfate added (100, 400 and 500 mg of $CuSO_4$ in 100 mL of clinical gel). Samples were photographed before and after exposure and color change evaluated with the ImageJ© program. It was observed color change in the samples as the radiation dose increased. Higher color variations were observed for the sample of clinical gel/$CuSO_4$ (100 mg). The samples were re-evaluated one month after irradiation and it was noticed that the color undergoes alteration over time, but without returning to the original color. These results are useful in characterizing future use of the composite as a volumetric indicator of dose distribution in gamma irradiation processes.

Keywords: **Clinical gel, $CuSO_4$, gamma irradiation process.**

ISSN: 2319-0612
Accepted: 2020-11-23

1. INTRODUCTION

The ability of the visual accompaniment of the dose distribution in a given material irradiated with gamma radiation source can be a strategic tool for daily process control [1]. The radiochromic film dosimetry system provides a means for measuring absorbed dose based on radiation-induced change in color using spectrophotometers, densitometers or scanned images. Commonly they are used in industrial radiation processing, for example in the sterilization of medical devices and the irradiation of foods [2].

However a volumetric evaluation of the dose distribution cannot be performed with films, adapting to the gel, as proposed by Abdel-Fattah, et al. (2017), for low doses of gamma irradiation [3]. This is because the color change may not occur evenly throughout the dosimeter volume, showing a difference in dose distribution. This difference can be assessed by taking measurements at different points in the samples by a photocolorymeter or a spectrophotometer.

The $CuSO_4.5H_2O$ has already been studied to form part of the composition of gel dosimeter [4, 5], thus dosimeters are based on the dose measurement related to the effect of radiation on the polymeric structure of the gel [6]. The chemical process of color change in the materials induced by radiation is normally time-dependent interaction mechanisms, that should be understood for delimiting the reading time and signal fading.

In this sense, in the present work we propose color change of clinical gel/$CuSO_4$, by gamma radiation, samples were irradiated with doses between 10-100 kGy and evaluated with ImageJ© software, to quantify the variation in the blue value in the sample. The understanding of the transformations undergone by the compound is performed through Ultraviolet-visible spectroscopy (UV-Vis). Analysis of signal fading was performed in 10 and 30 days after irradiation.

In this work the gel dosimeter is studied for gamma irradiation of high doses, since the processes of gamma sterilization are also associated with the breaking of organisms with similar human body composition (C, H, O), like microorganisms [1]. Thus we see application in processes of sterilization of food and medical instruments, among others. The radioinduced color changes in of the gel are recorded using a CCD camera, and analyzed using the ImageJ© software.

2. MATERIALS AND METHODS

Samples of the $CuSO_4.5H_2O$ (100, 400 and 500 mg) in 100 mL of colorless clinical gel RMC® from Unigel manufacturer was prepared by manual mixing. Samples were irradiated in Gamma Irradiation Laboratory (LIG) of Centro de Desenvolvimento da Tecnologia Nuclear (CDTN) using a Co-60 source at constant dose rate (16,7 kGy. h^{-1}), for doses between 10 – 100 kGy in triplicate. Irradiation was performed in presence of air. The Fig 1(a) shows gel/$CuSO_4$ (100 mg/100 mL) sample to exemplify how the samples were prepared and (b) the samples in the three proportions of $CuSO_4$ as prepared for the irradiation process.

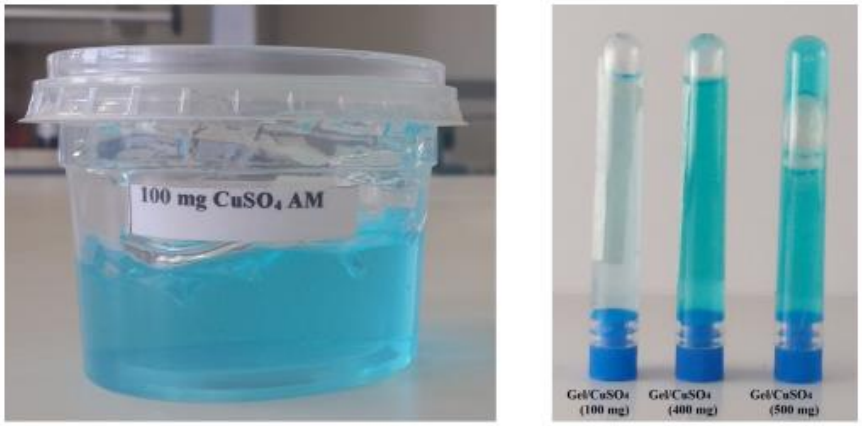

Figure 1: *Image of the compound in container used for mixing clinical gel and copper sulfate (a) and separate gel samples for irradiation with different proportions of copper sulfate (b).*

Images of samples were obtained using CCD camera for image processing and the digitized color was separated into RGB components with the ImageJ© software, enabling association between blue color intensities and absorbed dose. Optical transmission measurements were taken in a Shimadzu UV-2401 PC spectrometer, for wavelengths ranging from 200 to 800 nm. The measurements were performed after irradiation and repeated 10 and 30 days after.

3. RESULTS AND DISCUSSION

In Fig. 2 we present the results obtained by the analysis of the variation of the blue color of the gel/$CuSO_4$ (100 mg) samples. In Fig. 3 we present the results for the other samples, that is, gel/$CuSO_4$ (400 mg) in Fig. 3 (a) and gel/$CuSO_4$ (500 mg) in Fig. 3 (b). The figures show the responses analyzed by ImageJ© in terms of blue value against absorbed doses in the range of 10 kGy – 100 kGy. Clearly the blue values decrease proportionally with the increase of absorbed doses, although this proportionality is better perceived in the gel/$CuSO_4$ (100 mg) samples.

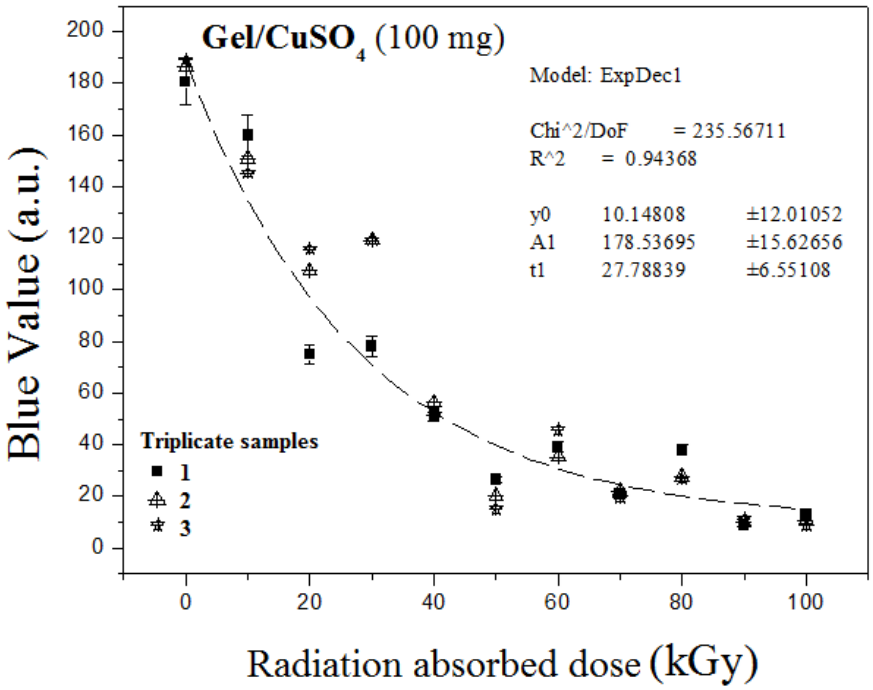

Figure 2: *Variation of blue values as a function of absorbed dose of the gel/$CuSO_4$ (100 mg) no irradiated and gamma irradiated with doses between 10 – 100 kGy.*

It can be observed that all responses have an exponential trend, but the best fitting shows the gel/$CuSO_4$ (100 mg) sample as the most adequate for dosimetric use. The other samples showed great dispersion in the values obtained, resulting in adjustments with inferior quality.

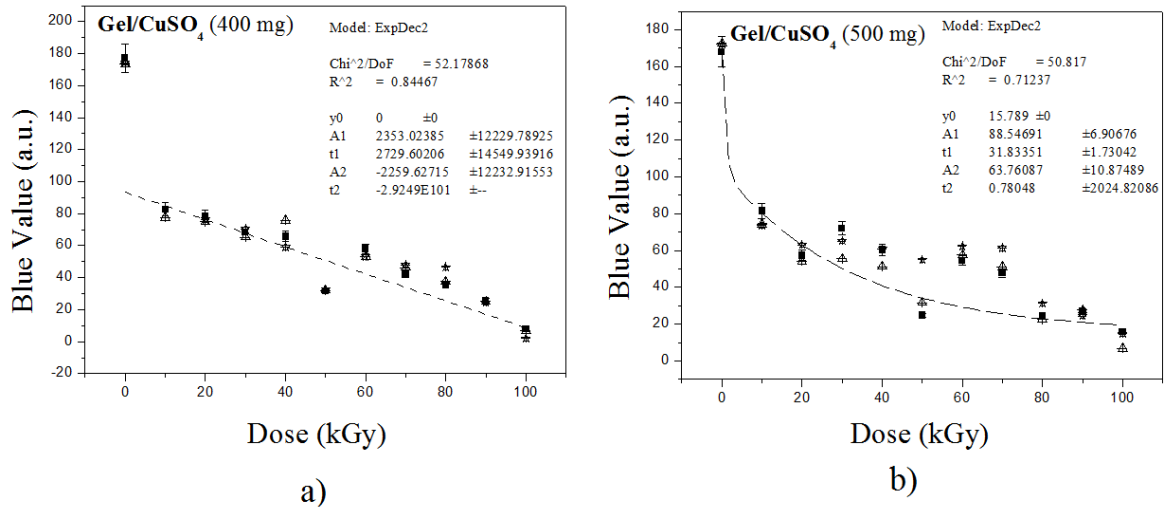

Figure 3: *Variation of blue values as a function of absorbed dose of the (a) gel/$CuSO_4$ (400 mg) and (b) gel/$CuSO_4$ (500 mg) samples no irradiated and gamma irradiated with doses between 10 – 100 kGy.*

For comparison of the effect of the radiation absorbed dose on the gel/$CuSO_4$ to explain the differences between the samples prepared with different proportions of copper sulfate, we present the UV-Vis spectra of the respective samples in Fig. 4. In Fig. 4 (b) the sample irradiated with 70 kGy was used to exemplify what also happened with the other samples (gel/$CuSO_4$ 400 and 500 mg), for a clearer visualization.

The UV-Vis spectra of gel/$CuSO_4$ no-irradiated sample show peak at 700 nm characteristic of the sulfate group. We observed in the spectra of the gel/$CuSO_4$ (100 mg) samples the appearance of a peak at 470 nm that rises as the absorbed dose increases. Peaks in this region are associated with the presence of CuO, which would be the result of the interaction of radiation with the samples [7,8].

Considering that for the dosimetric purpose the more marked change in the color of the sample with 100 mg of $CuSO_4$ is more strategic for a visual evaluation and for measurement of color variation, it is important to observe whether this change is permanent or temporary. Most of the chemical dosimeters used for high doses of gamma radiation have a limited time for dose measurement because post-irradiation chemical rearrangements.

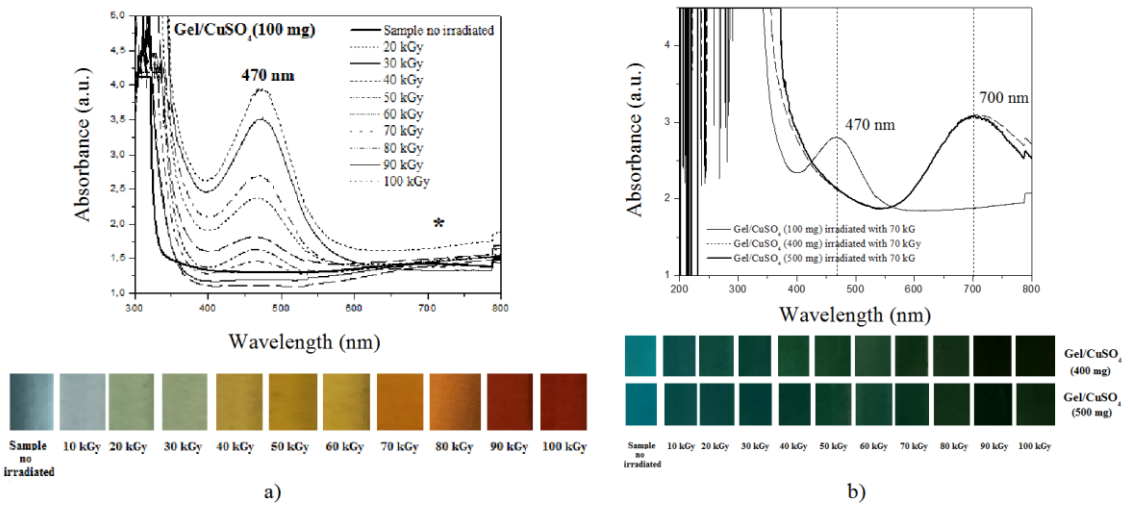

Figure 4: *UV-Vis spectra of gel/$CuSO_4$ (100 mg) non irradiated and gamma irradiated with doses between 10 – 100 kGy (a) and UV-Vis spectra of gel/$CuSO_4$ (100, 400 and 500 mg) irradiated with 70 kGy (b). The spectra were obtained shortly after irradiation. Images of the samples showing the color variation with the dose are below the UV-Vis spectra, for gel/$CuSO_4$ (100 mg) in (a) and for gel/$CuSO_4$ (400 and 500 mg) in (b).*

The UV-Vis spectra of gel/$CuSO_4$ no-irradiated sample show peak at 700 nm characteristic of the sulfate group. We observed in the spectra of the gel/$CuSO_4$ (100 mg) samples the appearance of a peak at 470 nm that rises as the absorbed dose increases. Peaks in this region are associated with the presence of CuO, which would be the result of the interaction of radiation with the samples [7,8]. The color of copper sulfate pentahydrate is blue and copper oxide is red, and in fact, we observed color change in samples irradiated from blue to red (Fig. 4a). Decomposition of copper sulfate into water forming copper oxide (described below) can occurs in two steps, but is not readily achievable.

$$CuSO_4.5H_2O(s)\ CuSO_4(s) + 5H_2O(l) \quad (1)$$

$$CuSO_4(s)\ CuO(s) + SO_3(g) \quad (2)$$

Research groups have investigated the hybrid copper oxide – copper sulfate cycle for the thermochemical splitting of water for hydrogen production and report the need for high

temperatures and pressure control. In fact for the decomposition predicted in Reaction 1 would require a temperature of 208 °C and for Reaction 2 require 850 °C [9]. We consider that the high energy deposited by gamma radiation facilitates the process and the initial concentration of copper sulfate seems to determine its efficiency.

For samples with the highest $CuSO_4$ proportion did not observe the same effect because the color variation occurs in blue and green tones (Figure 4b). In the UV-Vis spectrum of the gel/$CuSO_4$ (400 and 500 mg) samples we observed that the original peak at 700 nm remains visible and increases in intensity with increasing dose, which is shown in Fig. 4b with the spectrum of samples irradiated with 70 kGy. Different from the sample with 100 mg of $CuSO_4$ in which we observed this increase of absorption at the peak at 470 nm.

In Fig 5 we show the UV-Vis spectra of the gel/$CuCO_4$ samples (100 mg) irradiated with 100 kGy.

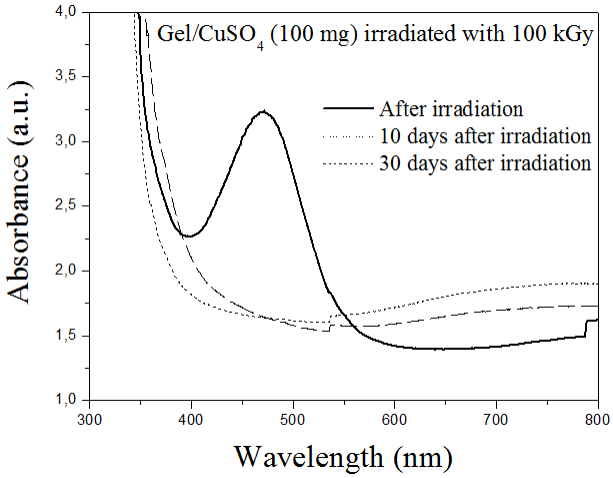

Figure 5: *UV-Vis spectra of gel/$CuSO_4$ (100 mg) gamma irradiated with dose of the 100 kGy, measurements performed shortly after irradiation, 10 and 30 days later.*

Measurement made shortly after irradiation, others referring to measurements performed 10 and 30 days after irradiation. We observed that the peak centered at 470 nm that appears in the spectrum of the measurement performed shortly after irradiation disappears in the others spectra. This demonstrates that rearrangements occur in the sample after irradiation, reversing its effect. Thus it is foreseeable that the sample also changes color, returning to the characteristic blue tones of $CuSO_4$.

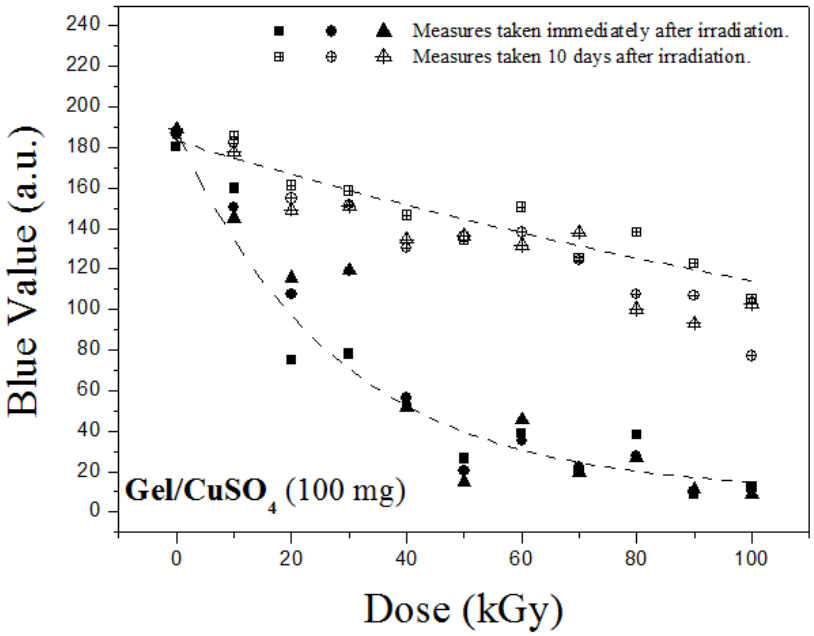

Figure 6: *Variation of blue values as a function of absorbed dose of the gel/$CuSO_4$ (100 mg) no irradiated and gamma irradiated with doses between 10 – 100 kGy after irradiation and 10 days after irradiation.*

We can observe a linear behavior with little variation between the blue values in the samples when measured 10 days after the irradiation, thus suggesting the need to carry out the measurements soon after the irradiation to obtain a good calibration curve. Immediate measurement is feasible even in an industrial installation, since sophisticated equipment is not required.

4. CONCLUSION

The gel/$CuSO_4$ samples prepared with 100 mg de $CuSO_4$ in 100 mL of the gel undergoes visual color change after exposited at high gamma dose (10 – 100 kGy) from blue into green–red by radiation induced decomposition of copper sulfate and subsequent appearance of copper oxide by interaction with the gel. Gel samples with other $CuSO_4$ proportions (400 and 500 mg) did not exhibit the same effect, showing less expressive color variation, into blue–green. In gel/$CuSO_4$ (100 mg) its blue–red coloration appears upon irradiation to 10 kGy confirming the capability of use as a

radiation yes/no dosimeter, for irradiation visual check, in gamma irradiation process. Evaluation performed 10 and 30 days after irradiation demonstrates the need to perform dose measurements soon after irradiation. The measurements performed in this work were carried out under the same conditions of the routine irradiations, thus, at the usual working temperature in the irradiator. Different conditions, such as those that provide greater protection of the sample as to temperature and dose rate variation, may be evaluated in future work.

ACKNOWLEDGMENT

Financial support from Conselho Nacional de Desenvolvimento Científico e Tecnológico (CNPq) and Fundação de Amparo a Pesquisa do Estado de Minas Gerais (FAPEMIG) are acknowledged.

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