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Development and characterization of polymeric composite material for use as radiation dosimeter

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Abstract

A polymer-based composite material was developed for use as a yes/no indicator of the high-doses gamma irradiation process. It was prepared with glycerin, gelatin, formaldehyde, clinical gel and copper sulfate pentahydrate (0.1% in relation to the total mass of the composite). Copper sulfate was intended as a chemical indicator of radiation exposure, with mechanisms focused on the processes of interaction with radiation. The formation of copper oxide and sulfur compounds was predicted, which would result in a color change, since $CuSO_4.5H_2O$ confers a blue coloration to the composite. To evaluate the role of copper sulfate in the production of a chemical indicator of radiation exposure, a sample without $CuSO_4.5H_2O$ was also prepared. The samples were irradiated with doses of 50, 80 and 100 kGy of gamma radiation from a Cobalt-60 source, in the Gamma Irradiation Laboratory of the Nuclear Technology Development Center, which has a multipurpose panoramic irradiator. The samples were characterized by Fourier-transform infrared spectroscopy (FTIR), demonstrating their glycerin-based composition. To evaluate the color variation in the irradiated and non-irradiated samples, they were photographed and analyzed using the color catalog of the Adobe Color® application. The sample with $CuSO_4.5H_2O$ showed significant color variation after irradiation, ranging from shades of blue to red/brown. The results demonstrate the potential of polymer composite whit sulfate pentahydrate for use as yes/no indicators of the irradiation process. It encourages future work to evaluate the composite for dosimetry purposes.

Keywords: Materials composites; Polymer; Dosimetry.

1.- INTRODUCTION

Dosimetry is a key field in radiation science and technology, with applications ranging from radiation protection to monitoring industrial and medical processes. One promising approach in this area involves the use of modified polymer composite materials to act as dosimeters, enabling the detection and quantification of ionizing radiation, such as gamma radiation [More *et al*., 2021; Zeng *et al.,* 2023; Adliene *et al.,* 2020].

Copper sulfate ($CuSO_4.5H_2O$), a compound widely used in various industrial and laboratory areas, has interesting properties when incorporated into polymer matrices [Liu *et al.,* 2016; Purcea Lopes *et al*., 2022]. In addition to giving the material a characteristic blue color, $CuSO_4.5H_2O$ can undergo chemical transformations under irradiation, such as, for example, color change [Yamagushi *et al.,* 2016; Kostyuk and Dick 2024]. This color change indicates a potential application of the polymer composite as a visual dosimeter or as a binary (yes/no) signal for high-dose gamma radiation irradiation processes.

Previous studies suggest that ionizing radiation can induce chemical reactions in copper-containing materials, promoting structural changes that affect their optical and chemical properties [Yamagushi *et al.,* 2016; Kostyuk and Dick 2024]. These changes are of particular interest for the development of new radiation-sensitive materials that can provide rapid and visible responses to the irradiated environment. Thus, the investigation of the irradiation of polymer composites containing copper sulfate may open new avenues for the creation of simple and efficient dosimetry devices, with application in areas where accurate and reliable radiation detection is critical.

In this context, the present work aims to evaluate the effect of high-dose gamma radiation on a polymer composite added with copper sulfate, focusing on the analysis of the possible chemical transformation of $CuSO_4.5H_20$, and the consequent color change from blue to red.

The objective is to explore the potential of this material as a radiation dosimeter or as a visual indicator of gamma irradiation in industrial processes.

2.- MATERIALS AND METHODS

The composition of the polymeric base material used in this research involved the addition of copper sulfate pentahydrate (0.1% $CuSO_4.5H_2O$ in relation to the total mass of the composite) in a mixture of glycerin, gelatin, water and formaldehyde. The samples were molded in round containers (150x150 mm petri dish). Figure 1 shows the appearance of the sample with and without copper sulfate.

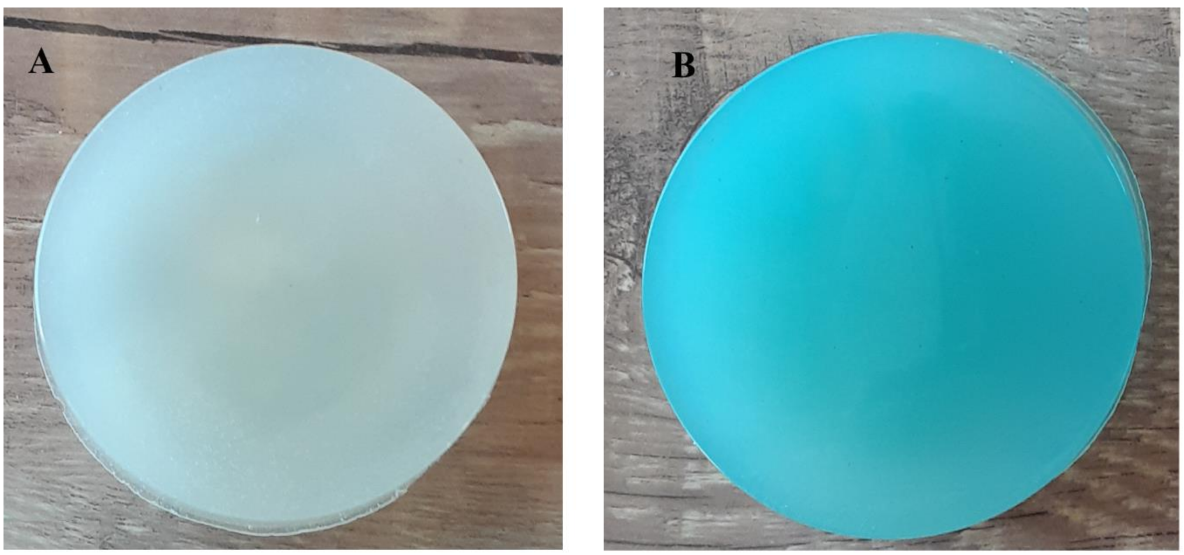

Figure 1.- In A image of the sample prepared with glycerin, gelatin, water and formaldehyde; in B, sample with the addition of copper sulfate pentahydrate in clinical gel mixture.

The preparation of the samples followed the following procedures: the gelatin was diluted in water and heated over low heat until it reached a homogeneous consistency. The other materials were added, except for the formaldehyde, mixing gently and removing the foam that formed. Finally, formaldehyde was added and mixed gently. The mixture was poured into molds, left at room temperature for up to 12 hours, then unmolded and stored in plastic wrap. Fourier Transform Infrared Spectroscopy (FTIR) spectra were obtained on a Thermo Scientific equipment model Nicolet 6700, with 64 scans ranging from 4000 to 650 cm^{-1} at a resolution of 4 cm^{-1}.

The samples were irradiated with doses of 50, 80 and 100 kGy of gamma radiation from a Cobalt-60 source at the Gamma Irradiation Laboratory of the Nuclear Technology Development Center, which has a multipurpose panoramic irradiator. To assess the color variation in the irradiated and non-irradiated samples, they were always photographed under the same lighting conditions, using a 13-megapixel camera, with a resolution of 4163 x 3122 pixels, and analyzed using the color catalog of the Adobe Color® application. Through the application, it was possible to extract the colors present in the image of the samples, forming a color palette in the RBG (Red, Blue, Green) pattern, where three primary colors (red, green and blue) are combined to compose a large set of colors within the color range of human vision [Santos and Pereira 2013]. The color palettes generated were evaluated for the variation in the chromaticity of the samples, illustrated in a diagram obtained from combinations of more saturated colors. Thus, it is possible to assess the extent of the color variation by observing the distribution of the points in the diagram.

3.- RESULTS

The samples were evaluated by FTIR to characterize the material. The results can be seen in Figure 2.

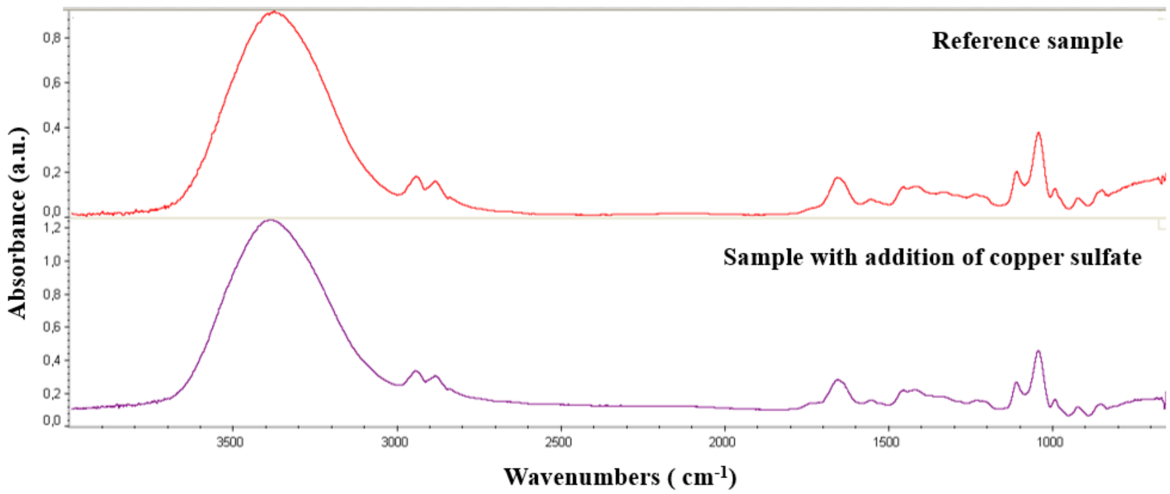

Figure 2.- FTIR spectra of the reference sample and of a sample with addition of copper sulfate pentahydrate in clinical gel mixture.

Figure 3 shows the composite samples containing $CuSO_4.5H_2O$ and the reference sample, before and after irradiation.

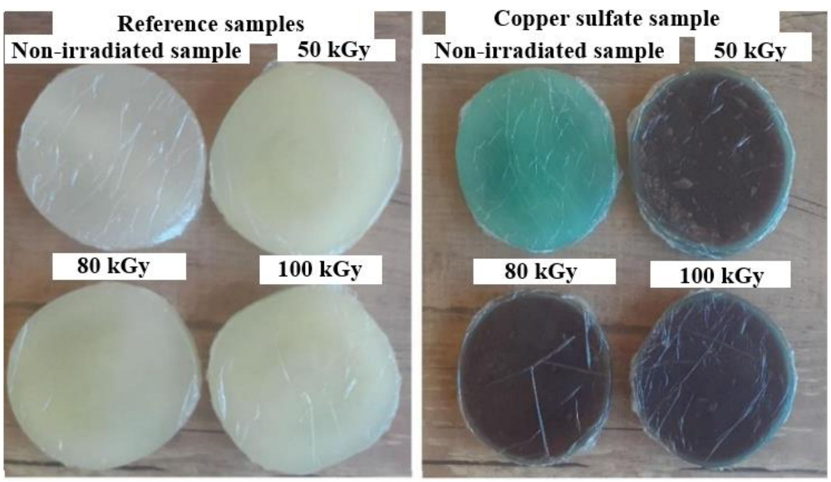

Figure 3.- Photo of non-irradiated and irradiated samples with doses of 50, 80 and 100 kGy.

To study the color variation of the samples before and after irradiation, they were analyzed using the Adobe Color® application. The results obtained are shown in Figure 4.

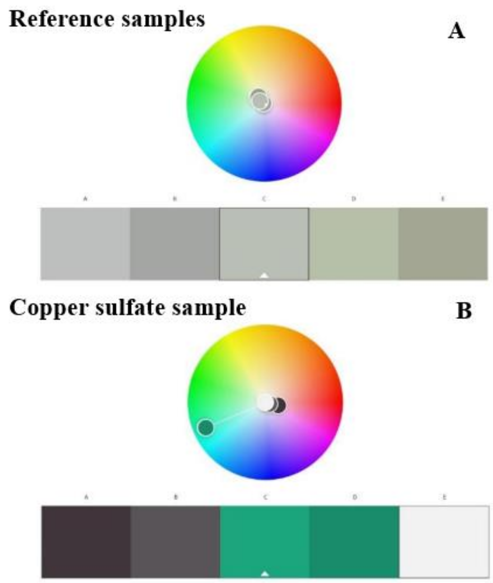

Figure 4.- Diagram and color palette in the RBG pattern of the samples before and after irradiation in A) sample without copper sulfate and B) sample with addition of copper sulfate.

Samples containing copper sulfate were reevaluated six months after irradiation, the photos are shown in Figure 5.

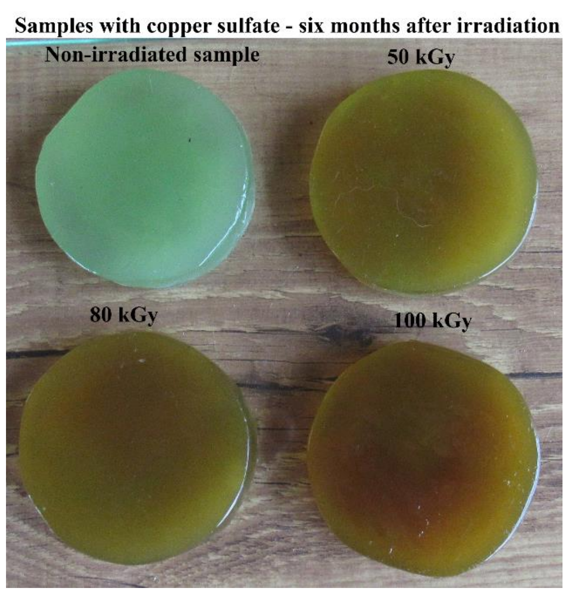

Figure 5.- Photo of non-irradiated and irradiated samples six months after irradiation.

4.- DISCUSSION

An analysis performed using the FTIR spectra in comparison with data from a database indicates the composition of the sample: predominance of glycerin, chemical formule $C_3H_8O_3$. The glycerin peaks in the spectrum prevent the visualization of other elements known to be present, such as copper sulfate itself, hence the similarity between the spectra. Therefore, the FTIR technique does not help in elucidating the mechanisms that lead to the color change caused by the irradiation process.

In visual analysis it is possible to observe that the samples with the addition of copper sulfate change color after irradiation. At the lowest dose (50 kGy), it is noted that the irradiation causes a color change in the samples to which copper sulfate was added. There is no noticeable change in the sample without copper sulfate, which demonstrates that $CuSO_4.5H_2O$ plays a decisive role in the process.

The samples were evaluated to compose a color diagram. Through the color palettes and signaling in the color diagrams obtained by the application, it was possible to confirm that there was greater radioinduced color variation in the sample with copper sulfate. In the sample to which copper sulfate was not added, there was practically no change in color, remaining concentrated in the central region of the color diagram.

For dosimetric use, it would be ideal if the color change caused by the interaction of radiation with the material were progressive, creating a spectrum of colors to be associated with the absorbed dose. However, a significant but abrupt change in colors was observed. This demonstrates its application as an indicator of irradiation. To this end, the samples need to be evaluated immediately after irradiation, since it was possible to observe new changes in color over time. This shows that there is a fading of the generated signal, although the sample does not return to its initial appearance.

Future work should be carried out by irradiating the material with lower doses. In this sense, doses above 50 kGy could already be considered a signal saturation for a dosimetric system. Another alternative is to reduce the dose rate, to study the material for high gamma dose irradiation, but deposited more progressively. Thus, the results indicate application of the composite material as an indicator of the irradiation process.

5.- CONCLUSIONS

A polymeric-based material was used to produce a composite with the addition of $CuSO_4.5H_2O$ to be used as an indicator of exposure to ionizing radiation. Reference samples and samples containing $CuSO_4.5H_2O$ were irradiated with doses of 50, 80 and 100 kGy. A color change was observed in the irradiated samples. The reference samples did not show any color change, making the role of copper sulfate in the process evident. The results are promising and encourage future work, irradiating the material at lower doses or lower dose rates. It is expected that, in this way, the color change will occur more progressively, favoring the use of the material as a dosimeter.

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