Radiation Physics and Chemistry 156 (2019) 307–313

Contents lists available at ScienceDirect

Radiation Physics and Chemistry

journal homepage: www.elsevier.com/locate/radphyschem

The influence of gamma irradiation on the color change of wool, linen, silk, and cotton fabrics used in cultural heritage artifacts

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ARTICLE INFO

Keywords:
Gamma irradiation
Fabrics
Color
Cultural heritage

ABSTRACT

Fabrics made of wool, linen, silk, and cotton, which are used in the making of artifacts of cultural heritage, were divided into three groups. The first group wasn't dyed, the second group was dyed with natural dye (using sour cherry), and the third one was dyed with artificial aniline dye. For the conservation purposes, all samples were exposed at ambient temperature to gamma radiation doses, from 0.5 to 25 kGy, using a Co-60 irradiator. The influence of gamma irradiation treatment on the color changes was investigated. The obtained results revealed that the irradiation affects the color changes in all samples. All samples became more or less darker under the influence of gamma irradiation. This change in darkness is most pronounced in samples dyed with natural colors. Also, the highest differences in color change and darkness were observed in linen samples.

1. Introduction

Cultural heritage (CH) occupies a prominent place in the culture and tradition of every nation and its role in history is very important as a symbol of ethnic identity, as emphasized in visual and aesthetic CH artifacts value. CH artifacts are exposed to the deteriorating effects of physical, chemical, and biological factors (Bethencourt et al., 2018). Among all organisms, microorganisms (bacteria, archaea, and fungi) (Sterflinger and Piñar, 2013), insect pests and lichens (Bates et al., 2011), have substantial biodeteriorationpotential, causing severe damage in the conservation of CH and health problems for restaurateurs and archivists. CH and art objects may be composed of a wide variety of materials (Bordet, 2018), both inorganic and organic, and may be subjected to a microbial attack that could result in their aesthetic and structural damage. Therefore, more complete information and valuable suggestions on the best conservation and preserving strategies are desirable (Laurila-Pant et al., 2015), since saving the CH has become a major concern of many modern societies. An important part of the CH artifacts is composed of textile materials, such as national costumes, uniforms, flags, carpets, and many other items (Mentges and Shamukhitdinova, 2017). Textile materials of wool, linen, silk, and cotton are mostly used and are susceptible to degradation. The use of gamma irradiation in order to decontaminate textile materials is a well-known technique (Hanh et al., 2014; Bashar et al., 2015; Drábková et al., 2018; Kavkler et al., 2018). However, in order to apply this technique to textile items belonging to CH, it is necessary to conduct detailed investigations of the influence of high-energy ionizing gamma radiation on textile materials, such as effects on their color changes, in order to extend their shelf-life of this kind of artifacts (Rehman et al., 2013; M'Garrech and Ncib, 2009).

The recovery of textile materials infected by living organisms can be performed by gamma ionizing radiation, with many advantages over other sterilization or disinfection methods (Takács et al., 1999). Gamma radiation does not leave toxic or radioactive residues inside the treated items or in the environment and is a penetrating contactless method (Leemhorst, 1995). Prior to routine application in conservation, the use of gamma radiation for CH artifacts (CHA) disinfection requires firmly established conclusions that irradiation does not lead to unacceptable changes (side-effects) in the functional or decorative properties of CH items. In this way, a method can be established to ensure a safer and more functional preservation of CH textile materials (Cortella et al., 2009).

Preserving art and cultural heritage is a shared ambition of the global community (Uses of Ionizing Radiation for Tangible Cultural Heritage Conservation, 2017). One of the most efficient ways is the application of radiation technologies for the characterization and preservation of cultural heritage. The preservation of CH artifacts (CHA) by using ionizing radiation, applied since the seventies of the last century, is increasingly being used in recent years. The irradiation of CHA in addition to its effectiveness in the decontamination is also a very reliable and safe method without leaving any harmful products for the health and safety of people.

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https://doi.org/10.1016/j.radphyschem.2018.12.001
Received 6 August 2018; Received in revised form 26 October 2018; Accepted 1 December 2018
Available online 04 December 2018

Different gamma radiation absorbed doses are needed for the destruction of organisms that can damage cultural heritage objects: a dose of 0.5–2 kGy is required for the destruction of the insects, 4–10 kGy for fungi and 5–25 kGy for complete decontamination (Machnowski et al., 2012a). Ionizing radiation can induce a modification in treated materials (sideeffects), occurring under irradiation as well as after the end of the radiation processing, such as loss of mechanical strength, optical, color and chemical changes (Baccaro and Cemmi, 2017).

In this paper, the irradiation tests have been performed at different absorbed doses (up to 25 kGy), carried out on samples before and after the irradiation treatments have been focused on optical properties of textile materials. The experimental measurements have been performed by spectrometric techniques.

Several studies on the effect of gamma irradiation on the change of fabric color have been performed. Chirila et al. have examined the influence of gamma irradiation on natural dyeing properties of cotton and linen fabrics. They found that irradiation positively influences the adsorption capacity of natural dye in cellulosic fibers, cotton and flax, irradiated with doses of 10, 25 and 40 kGy, leading to darker colors compared with the unirradiated samples (Chirila et al., 2018). Bhatti et al. have described the effect of gamma irradiation on the dyeing of cotton with an extract of turmeric. They concluded that gamma irradiation has not only increased the color strength of cotton fabric but also minimized the mordant concentration with improved fastness properties (Bhatti et al., 2010). Kavkler and collaborators have published that in non-aged contemporary silk gamma-irradiation to an absorbed dose of 6 kGy caused no color change compared to the non-treated sample (Kavkler et al., 2018). In this paper, we have examined the effects of different doses of gamma irradiation, up to 25 kGy, on several types of different textile materials. A comparative overview of color change depending on the type of textile is shown. Also, a comparison of the color change in undyed fabrics was made in relation to fabrics dyed with natural or artificial colors.

The effects of radiation depend on the absorbed dose, which must be large enough to effectively reduce the contamination of the fabric, but small enough not to damage the textile in any way. The results that were obtained in this work allow revealing the possible color changes in textile materials when treated with ionizing radiation. These results will provide a better understanding of the impact of gamma radiation on fabrics. In this way, it will be known in advance which doses are acceptable for the treatment of various textile materials in order to conserve and preserve CHA. By establishing such a method of preserving CH textile artifacts, it will be possible to apply this method to numerous specific objects, protecting and extending the shelf-life of textile items.

2. Materials and methods

2.1. Test material and samples

In order to analyze the effects of gamma radiation on the change in the color of the fabric, the patterns of wool, linen, silk, and cotton are irradiated in gamma facility with doses of 0.5, 1, 2, 3, 5, 7, 10, 15, 20 and 25 kGy. We examined the color change of undyed samples, as well as samples dyed with natural (sour cherry) colors, and artificial (aniline) colors.

2.1.1. Dyeing the fabric with natural color

Not all natural materials will produce a dye, and some produce colors that are nothing like the original plant it came from (Samanta and Konar, 2010). The most commonly used plants for natural fabric dyeing are carrots, onion skins (orange color), dandelion roots, oak bark, tea, coffee (brown color), berries, cherries, red and pink roses (pink color), red cabbage, blueberries, purple grapes (blue color), artichokes, spinach, peppermint leaves, peach leaves (green color) (Křížová, 2015).

Within this project, we dyed the fabric in a pinkish color with sour cherry. To prepare the fabric for dyeing, we put 5 g of salt in 4 cups water and added fabric. Then we brought the mixture to a boil, and let it simmer for about an hour. This creates a fixative that will help the fabric hold the color better, and which helps set the color in the fabric. Once finished, we rinsed the fabric in cold water and squeeze the excess water from it. For dyeing we put 1 cup of fruit and 4 cups of water in the saucepan. We brought the water to the boil, then added the fabric. The fabric was simmered gently for 30 min. Then we took the saucepan off the heat and put it, with the fabric and the dyed water still in it, aside to cool. Once the water is cooled we took the fabrics out. The longer the fabric is left in the water, the more of the color it will take on (Křížová, 2015).

2.1.2. Dyeing the fabric with artificial (aniline) color

Up until the mid-19th century, almost all dyes were made from materials found in plants. Then, in 1856 William Henry Perkin, a young chemistry student invented aniline dyes for the fabric (Holme, 2006). Aniline was one of the first synthetic dyes ever produced and was the first commercially successful synthetic dye (Garfield, 2008). Although aniline is rarely used as a dye today, artifacts of the cultural heritage are very often dyeing exactly by this kind of color. Most aniline dyes are available in dry powder or crystal form, from which the actual painting colors are mixed. One generally starts by mixing a concentrate of the color with the fabric and thinning down with water to get desired shade and amount. For the purpose of dyeing the fabric, we added about one quart of boiling water to three heaping teaspoons of dry dye that has been carefully placed in a glass or plastic container to minimize dust creation.

After dyeing, all the samples were individually packaged in plastic bags and irradiated with various doses of gamma radiation, as shown in Fig. 1.

2.2. Gamma irradiation

Irradiation was performed at a Co-60 γ-source of Radiation Unit for industrial sterilization and conservation, Vinca Institute for Nuclear Sciences, at room temperature, in contact with air, and the dose rate was about 10 kGy/h. Dosimetry measurements were performed by an ethanol-chlorobenzene solution (ECB) dosimetry (ISO/ASTM, 2009). The ECB solution is a well-known routine standard dosimeter for evaluating the absorbed dose of high-energy radiation (Kovács et al., 1987). Measurements were made by oscillotitrator OK-302/1 supplied by Radeliks Electrochemical Instruments, Budapest according to a calibration curve obtained at High Dose Reference Laboratory of Risø National Laboratory, Denmark.

2.3. Methods and analysis

The radiation-induced color change in studied nacres was monitored using CIE colorimetry, in the CIE Lab L*a*b* system. There measurements were performed with Spectrophotometer Shimadzu UV–Visible UV-2600 (Shimadzu Corporation, Tokyo, Japan) equipped with an integrated sphere (ISR-2600 Plus (for UV-2600)) in the 220–780 nm range and 1 nm step.

In order to analyze the results, the values ΔE* and ΔL* were calculated for each sample, where ΔE* represents the total color difference, and ΔL* - the difference in lightness and darkness (+ = lighter, - = darker). These values are calculated using the following equations (CIE, 2004):

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

$$\Delta L^* = L^*_{\text{irradiated sample}} - L^*_{\text{non-irradiated sample}}, \quad (2)$$

$$\Delta a^* = a^*_{\text{irradiated sample}} - a^*_{\text{non-irradiated sample}}, \quad (3)$$

$$\Delta b^* = b^*_{\text{irradiated sample}} - b^*_{\text{non-irradiated sample}}, \quad (4)$$

Fig. 1. samples of wool, linen, silk and cotton fabrics prepared for irradiation.

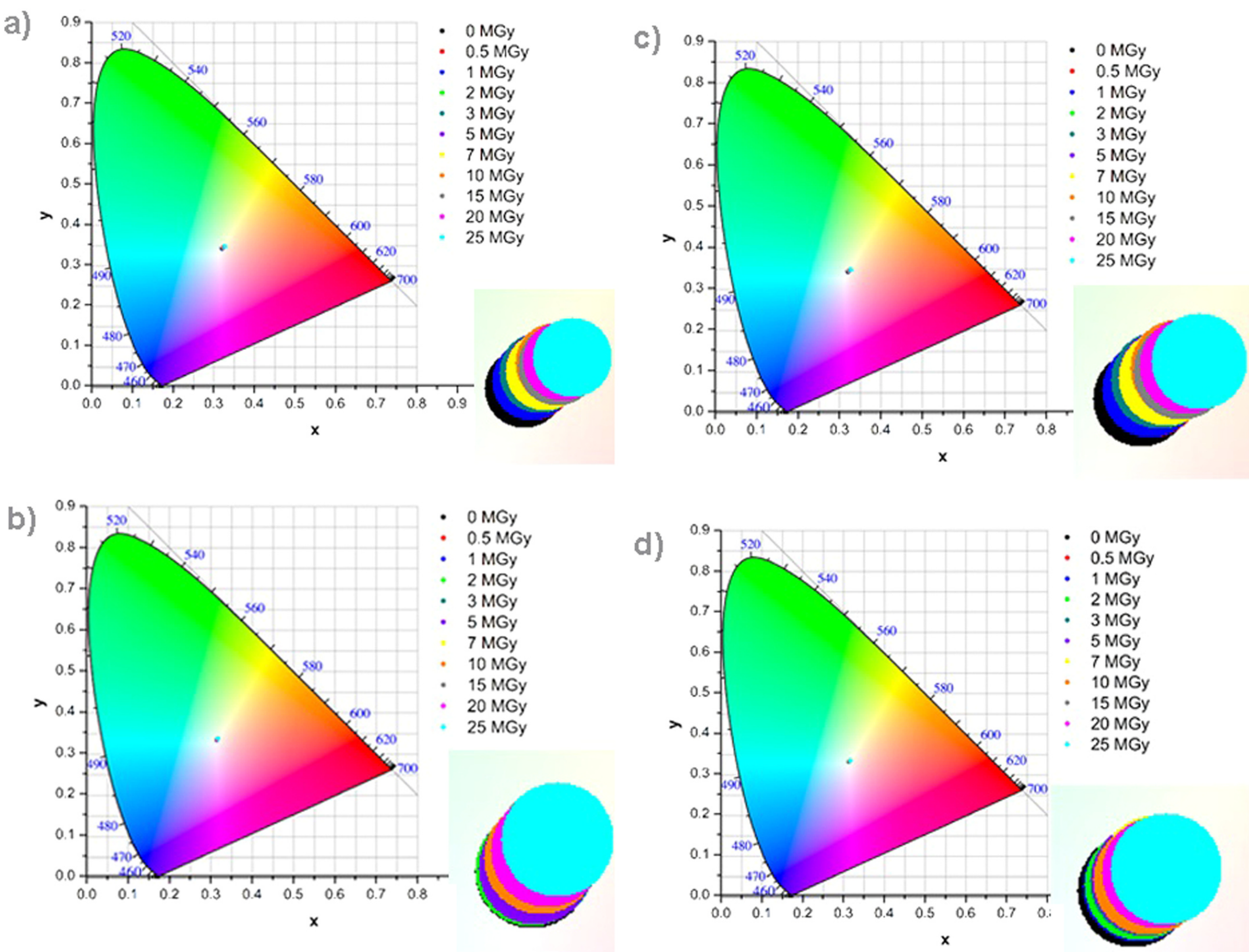

Fig. 2. The CIE chromaticity diagram of undyed samples of (a) wool, (b) linen, (c) cotton and (d) silk fabrics irradiated with different doses of gamma rays.

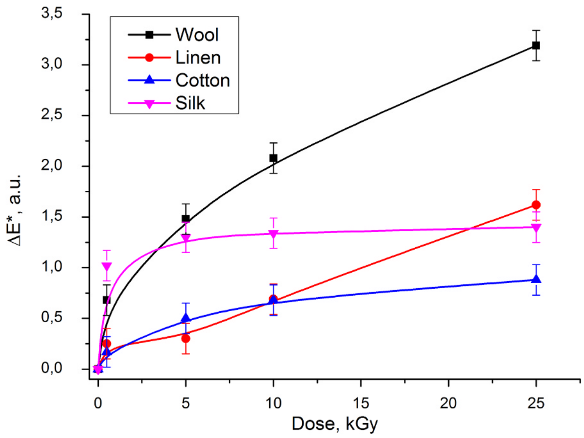

Fig. 3. Diagram of the total color difference for undyed fabric samples irradiated with different doses of gamma rays.

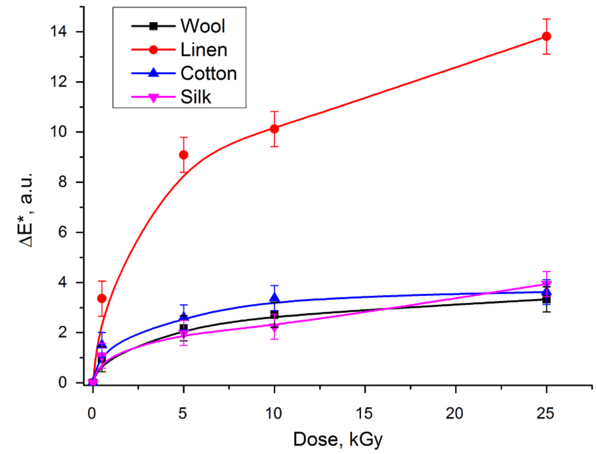

Fig. 5. Diagram of the total color difference for fabric samples dyed with natural colors and irradiated with different doses of gamma rays.

3. Results and discussions

3.1. Color changes in undyed samples

Firstly, we examined the color change in undyed wool, linen, cotton, and silk samples irradiated with doses of 0.5, 1, 2, 3, 5, 7, 10, 15, 20 and 25 kGy. Fig. 2 shows CIE diagrams for undyed samples of wool (a), linen (b), cotton (c), and silk (d) fabrics irradiated with different doses of gamma radiation. In the lower right corner of the image, an enlarged color distribution region is presented. From the figure, one can see that

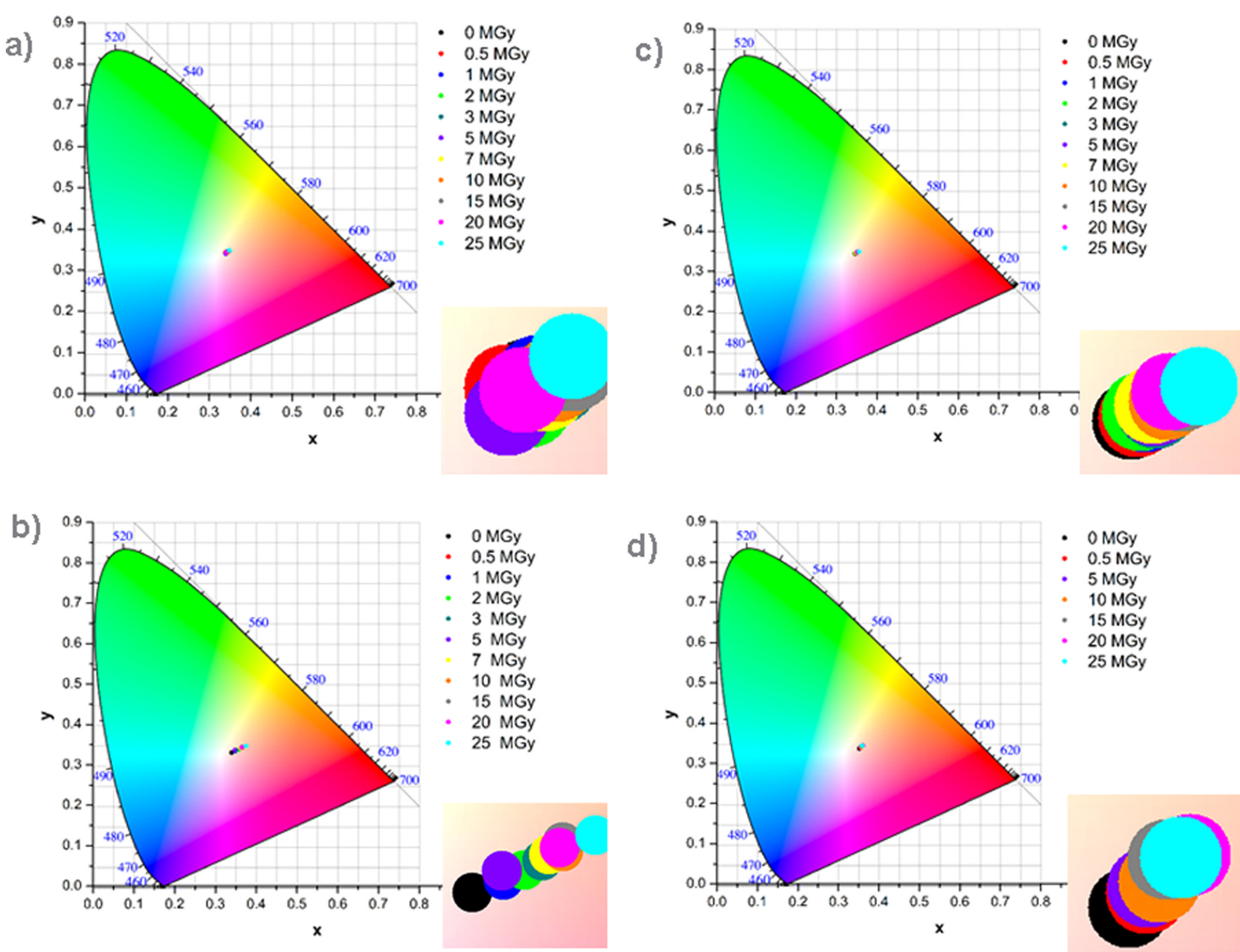

Fig. 4. The CIE chromaticity diagram of samples of (a) wool, (b) linen, (c) cotton, and (d) silk fabrics dyed with natural colors and irradiated with different doses of gamma rays.

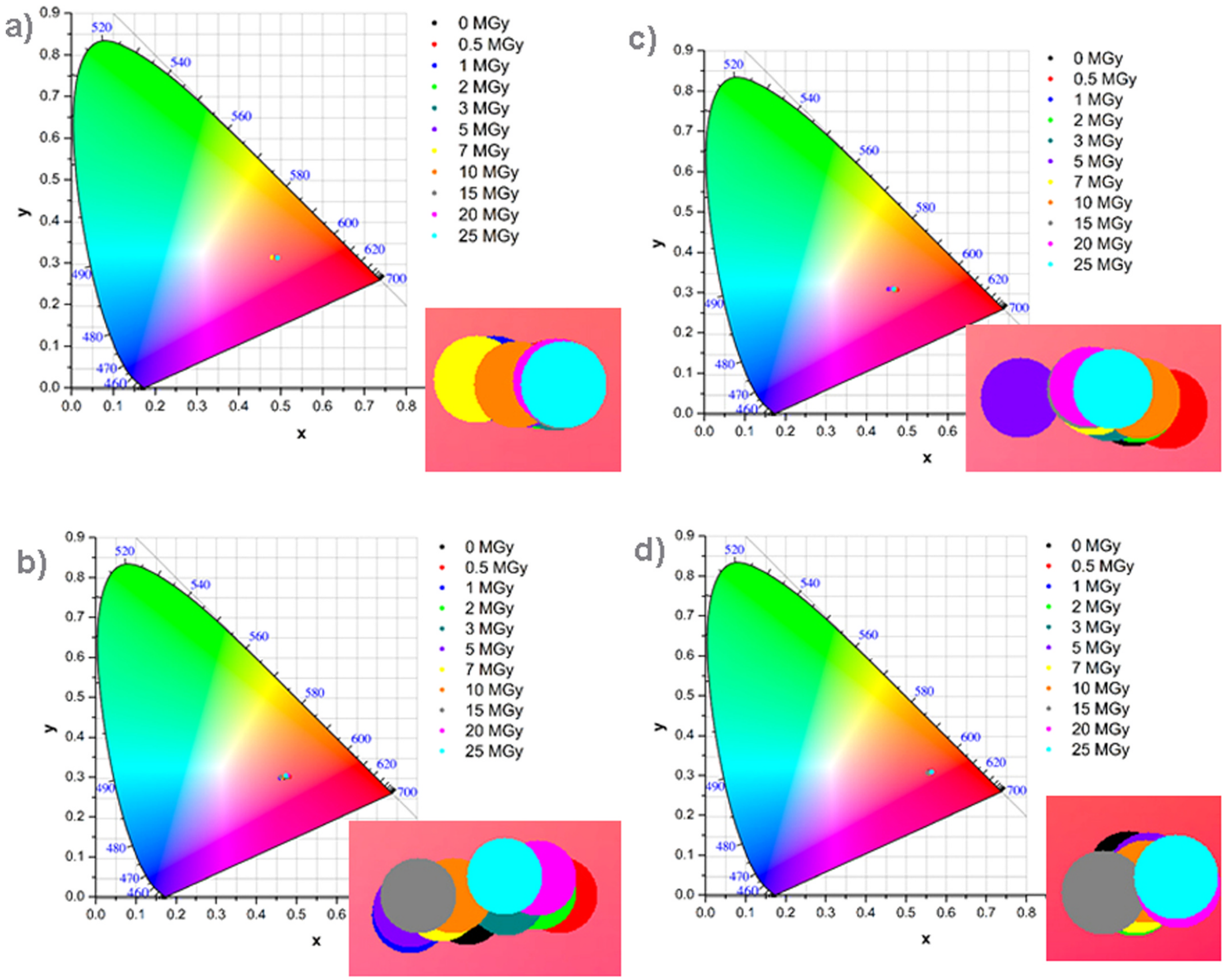

Fig. 6. The CIE chromaticity diagram of samples of (a) wool, (b) linen, (c) cotton and (d) silk fabrics dyed with artificial aniline colors and irradiated with different doses of gamma rays.

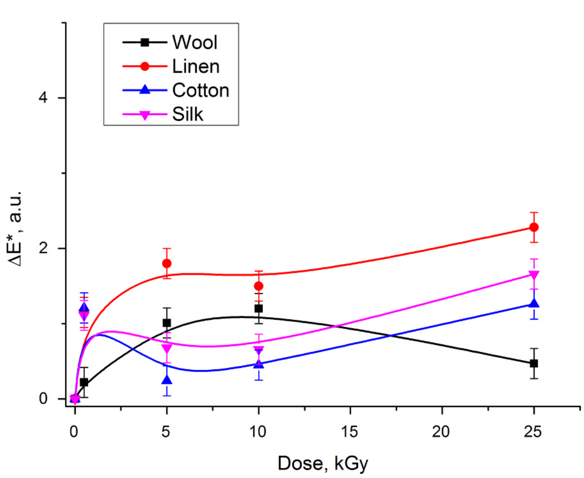

Fig. 7. Diagram of the total color difference for fabric samples dyed with artificial aniline colors and irradiated with different doses of gamma rays.

Table 1
The difference in lightness and darkness for samples of different fabrics irradiated with a dose of 25 kGy.

Undyed/Dyed	Fabric	$\Delta L^*_{25\,kGy}$	Average value
	Wool	− 1.38	
Undyed	Linen	− 0.17	− 0.60
	Cotton	− 0.74	
	Silk	− 0.10	
	Wool	− 2.79	
Dyed with natural colors	Linen	− 9.11	− 4.07
	Cotton	− 1.40	
	Silk	− 2.98	
	Wool	− 0.42	
Dyed with aniline colors	Linen	− 0.80	− 0.79
	Cotton	− 0.35	
	Silk	− 1.58	

the color change is almost linear with the increase in the dose. Also, as the dose increases, the samples become darker. On the diagram, black point represents non-irradiated sample and blue point represents 25 kGy irradiated sample.

The ΔE* value, which represents the total color difference, was calculated. Fig. 3 presents a diagram of the total color difference for fabric samples at different doses of radiation that they were exposed to.

From Fig. 3, one can see that the overall color change is almost linear for wool and linen fabrics, while for cotton and silk fabrics this change is significant at lower doses, and then almost does not change with increasing of irradiation dose. It is well known that wool yellowing due to several different factors such as sunlight, age, moisture, how it's stored (the environment), and irradiation, and this color change is conditioned by the specific composition of the wool (a high content of sulfur, a lot of suint, the natural grease, sweat, etc, due to the carotene in the corn that is feed or the darker grease that sheep can produce when it is exceptionally hot) (Duffield and Lewis, 1985). On the other hand, the color change in linen is conditioned by the high concentration of cellulose in the linen (Akin, 2013) that is sensitive to the influence of gamma radiation (Glegg and Kertesz, 1957). It should be noted that cotton, like linen, is a plant cellulose fiber too; however, these fibers clearly differ in anatomical structure. Cotton is a mono-cell fiber, while linen is a multicell stem fiber with noncellulosic impurities: hemicellulose, lignin, and pectin. It can be assumed that this considerable color change in linen fabric under the influence of gamma irradiation is due not only to the degradation of the small individual linen fibers but also to the degradation of some noncellulosic impurities, which bond these small individual fibers (Machnowski et al., 2012b).

Fig. 4 shows the color change of fiber samples dyed with natural color.

From the Fig. 4 it is noticed that the distribution of the color differences is not as linear as in the undyed samples. The reason is that the samples cannot be perfectly uniformly dyed, and there are small color deviations in the initial samples, so the color change distribution is not perfectly linear, but the diagram shows that color changes are much larger than in undyed samples. This can even be better seen in Fig. 5 that represents the total color difference. It can be noticed that the value of ΔE^* are much higher (the values of ΔE^* go up to 14)than in relation to undyed samples (the values are up to 3). The reason for this is the breakup of chemical bonds in organic molecules under the influence of gamma irradiation (Burton, 1947).

The largest color difference is observed in linen samples, due to the high content of cellulose and noncellulosic impurities as previously explained (Glegg and Kertesz, 1957; Machnowski et al., 2012b). For wool, cotton, and silk this change is significant at lower doses, and then almost does not change with increasing of irradiation dose.

Fig. 6 shows the CIE diagram for samples dyed with artificial aniline dyes. In this case, the change of color is very small, and the color change distribution is not followed by any pattern. The reason for such a small change in color is the high stability of aniline to the effect of ionizing radiation (Wolszczak et al., 1996).

Fig. 7 shows a very small total color difference, total value up to 2, practically negligible in comparison with natural dyes, which confirms the stability of aniline to the effect of gamma irradiation.

In order to determine the difference in lightness and darkness after irradiation, we calculated ΔL^* values for samples of different fabric irradiated with a dose of 25 kGy and the results are shown in Table 1.

The Table 1 shows the values of ΔL^* for each individual sample, as well as the average values for the undyed fabrics, fabrics dyed with natural color and fabrics dyed with aniline color. The negative values indicate an increase in the darkness of the samples, and positive values indicate an increase in lightness. One can see that all samples became more or less darker under the influence of gamma irradiation (all tested samples have negative ΔL^* values). However, this change in darkness is most pronounced in samples dyed with natural colors. It appears that irradiation with gamma rays influences the adsorption capacity of the natural dye in cellulosic fibers irradiated with the different doses of gamma irradiation, leading to darker colors, as described in the literature (Chirila et al., 2018).

4. Conclusions

In this paper, we examined the effect of gamma radiation on color changes of the fabrics used in the production of cultural heritage artifacts. Various types of fabrics (wool, linen, cotton, and silk) were exposed to a wide spectrum of ionizing gamma radiation (0.5, 1, 2, 3, 5, 7, 10, 15, 20 and 25 kGy).

The obtained results revealed that the irradiation influences the adsorption capacity of dye in wool, linen, cotton, and silk fibers, especially of natural dye, leading to darker colors compared with the unirradiated samples. All samples became more or less darker under the influence of gamma irradiation. This change in darkness is most pronounced in samples dyed with natural colors. This is caused by the particularly high absorption potential of the fruit used to dye the fabric.

Also, the highest differences in color change and darkness were observed in linen samples, which is conditioned by the high concentration of cellulose in the linen that is sensitive to the influence of gamma radiation.

It can be concluded that when using the technique of conserving the fabric by irradiation, it is very important to take into consideration origin of the dye with which fabrics were treated.

Acknowledgments

Authors acknowledge the financial support of the Ministry of Education, Science and Technological Development of the Republic of Serbia (Project No: 45020)

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