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Investigation of the response of chromium nitrate solutions as a chemical dosimeter for agricultural applications

Sedigheh Kashian[a], Hamideh Daneshvar[a,*], Peiman Rezaeian[a], Mohammad Rafiean[b]

[a] *Radiation Application Research School, Nuclear Science and Technology Research Institute, Tehran, Iran*
[b] *Islamic Azad University Science and Research Branch, Tehran, Iran*

HIGHLIGHTS

- Chromium Nitrate solutions with different concentrations have been prepared.
- Irradiation was performed with gamma rays.
- Dosimetery factors was invesitegitaed in the range of irradiation of agricultural products.

ABSTRACT

Irradiation of agricultural products is used to optimize properties and pest control and also increase the storage time of products. The desired irradiation result is obtained when the required dose is given to the sample. Dosimetry is a method for separating and classifying materials and equipment that is provided to the user to confirm the acceptance of irradiation and control the performance. In this work, Chromium Nitrate solutions with concentrations of 0.16, 0.24, and 0.32 mM have been prepared and irradiated with gamma rays between 100 to 1000 Gy. The purpose of this study is to investigate the dosimetry of these samples in the range of irradiation of agricultural products. Results show that the higher concentration sample is linear in about 100 to 1000 Gy dosimetry range and the optimal concentration must be found to achieve a stable sample in about 3 weeks periods. Also, samples that are in a darker environment are more stable than samples that are in a lighter environment.

KEYWORDS

Chromium nitrate
Chemical dosimeter
Agricultural products
Radiation processing

HISTORY

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1 Introduction

The use of ionizing radiation in various fields such as medicine, research, and industry is constantly expanding to achieve the final goal in radiation processing (Attix, 2008; Kojima et al., 1997). Industrial applications of radiation processing such as food irradiation, sterilization of health and medical materials, industrial wastewater treatment, prevention of germination and control of food insects, and processing of polymeric materials to improve physical and mechanical properties are done by using ionizing radiation (Fink and Rehmann, 1994; Sun and Chmielewski, 2017; Tauxe, 2001). The irradiation process has been extensively studied over the past 50 years. During these years, the effect of radiation on maintaining the quality of all types of food has been investigated and the limitations of the application of this method have been identified (Munir and Federighi, 2020). Irradiation of agricultural products is used to optimize properties, and pest control and also increase the storage time of products. The desired irradiation result is obtained when the required dose is given to the sample. If the amount of absorbed dose in the sample is less than the specified value, it will not be effective and if the amount of absorbed dose is higher, it could have destructive effects on the sample. Excessive doses cause changes in the taste, color, and smell of the product and may affect the nutritional components of the product such as moisture, sugar, and vitamin C. Therefore, measuring the absorbed dose in the samples has special importance. For this purpose, it is tried to use dosimetry systems with high accuracy and precision in measurement (McLaughlin and Desrosiers, 1995). Dose measurement provides a formal tool to confirm its use in sterilization of medical devices and food irradiation, where radiation can affect general health,.so dosimetry in radiation processing is one of the most important factors in achieving the goal (McLaughlin and Desrosiers, 1995).

*Corresponding author: hdaneshvar@aeoi.org.ir

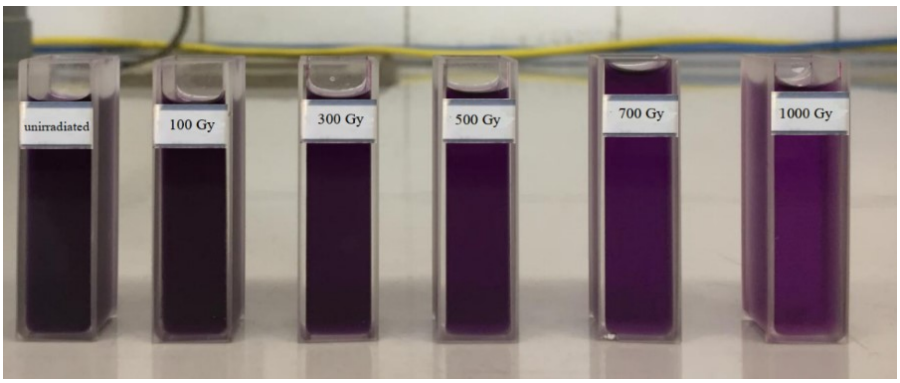

Figure 1: Irradiated sample of Cr/DPC liquid dosimeter with a concentration of 0.32 mmol.L^{-1}.

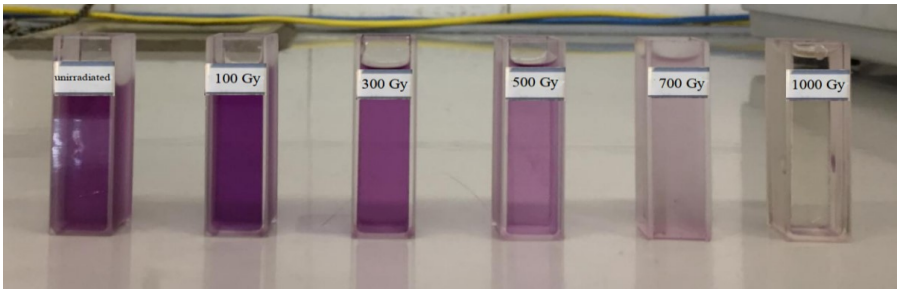

Figure 2: Irradiated sample of Cr/DPC liquid dosimeter with a concentration of 0.16 mmol.L^{-1}.

Dosimetry is a method for separating and classifying materials and equipment that is provided to the user to confirm the acceptance of irradiation and control the performance (Kojima et al., 1997; McLaughlin and Desrosiers, 1995). Different types of dosimeters have been used for this purpose, including gas ionization chambers, thin films, solid and liquid dosimeters (Cameron, 1963; McLaughlin and Desrosiers, 1995). Due to the dose ranges in the dosimetry of agricultural products, ie 10 to 1000 Gy, there are not many choices that cover the entire range (Adachi et al., 2004). It is not easy to obtain these dosimeters in the country because of high prices and very difficult access due to sanctions. Therefore, it is necessary to achieve a reliable, inexpensive dosimeter, easy manufacturing process, and comfortable reading system for use in the daily dosimetry of agricultural products (Adachi et al., 2004). Dosimeters used to measure cumulative doses are mainly divided into physical and chemical dosimeters. The most well-known type of chemical dosimeters is standard ferric dosimeters. The purpose of this study is to fabricate chemical dosimeters of base solution of Chromium Nitrate compounds to use in the desired dose range. Transition metals are intermediate elements whose d-layers are being filled (they have incomplete *d*-layers). Due to the presence of relatively large single electron orbitals in the d level and the possibility of proper overlap of these orbitals and the formation of covalent bonds in the crystal lattice, these elements tend to form complex color structures (Adachi et al., 2004). Radiation can break or weaken the covalent bond of the complex, and thus change its color. Much work has been done in this field in Iran and other countries (Abtahi et al., 2013; Edalatkhah et al., 2022; Gafar et al., 2018; Gorjifard and Sharifzadeh, 2003). This property can be used for the dosimetry process. In chemical dosimeters, the amount of absorbed dose is measured by quantitatively measuring the chemical changes caused by ionizing radiation. There are two stages in chemical dosimetry. The first stage is ionization due to the interaction of energetic charged particles that occurs at very high speeds (10^{-10} to 10^{-11} s) and the second stage is the chemical changes caused by radiation (Adachi et al., 2004). The purpose of this study is to fabricate soluble chemical dosimeters based on chromium compounds for use in the irradiation of agricultural products. The expected sensitivity for these dosimeters is in the range of doses used for irradiation of agricultural products. The color change can be read by changing the light absorption using a spectrophotometer. In this work, the characteristics of the dosimeter such as accuracy, linearity of the dosimeter response, and the effect of environmental factors such as temperature and light on the stability of the response before and after irradiation will be evaluated.

2 Experimental: Preparation of the materials

To prepare dosimetry solutions, Chromium Nitrate ($Cr(NO_3)_3$) and DPC (DiPhenylCarbazone), were used. Samples should be in concentrations of 0.32, 0.24, and 0.16 mM of Chromium Nitrate and 0.536, 0.402, and 0.268 mM of DPC, respectively, and in a volume ratio of 1 to 3

Table 1: Dosimetry equations of the Cr/DPC dosimetry solution samples.

Sample	Linear range of dosimetry response	Equation
0.16 mM	100 to 500 Gy $y = 0.0013\,x + 0.1302$ $R^2 = 0.9812$	$y = 0.2921\ln(x) - 1.0926$ $R^2 = 0.9835$
0.24 mM	300 to 1000 Gy $y = 0.0006\,x + 0.2209$ $R^2 = 0.9827$	$y = 0.2725\ln(x) - 1.1436$ $R^2 = 0.9800$
0.32 mM	100 to 1000 Gy $y = 0.0006\,x + 0.0569$ $R^2 = 0.9808$	$y = 0.0006x + 0.0569$ $R^2 = 0.9808$

$V.V^{-1}$ of water and acetone are mixed together. The materials were mixed in a volume of 100 mL and stirred for 3 hours using a magnetic stirrer. 25 ml of distilled water and 75 mL of acetone were used to prepare the mixture. Spectrophotometric properties of this solution have been investigated at 540 nm. In the Figs. 1 and 2, irradiated samples can be seen at concentrations of 0.32 $mmol.L^{-1}$ and 0.16 $mmol.L^{-1}$, respectively.

The samples were read using a BECKMAN Coulter-Du 800 spectrophotometer in the range of visible light wavelengths from 400 to 800 nm. The cuvettes with dimensions of $1.2 \times 1.2 \times 4.5$ mm^3 were used as a container.

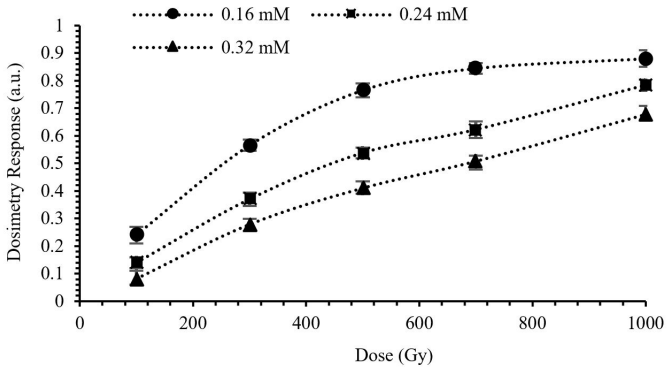

Figure 3: Dose-response curve of Cr/DPC liquid irradiated samples at different concentrations.

3 Results and Discussion

3.1 Dose response of liquid Chromium Nitrate

In this study, absorbance changes are plotted in terms of dose. To plot the dose-response curve, the following equation is considered:

$$Response\ dosimetry = \frac{HPUI - HPI}{HPUI} \qquad (1)$$

in this equation, $HPUI$ is the height peak of the unirradiated sample and HPI is the height peak of the irradiated sample (Gafar et al., 2018). The values of optical absorbance are measured at 540 nm. Dose-response curves in different concentrations of Chromium Nitrate on the first day of irradiation are shown in Fig. 3. Results show that samples with the lowest concentration are the most sensitive ones. This sample is saturated in about 800 Gy. Table 1 presents the dosimetric equations of the Cr/DPC solution samples. According to linear equations, samples with 0.24 mM and 0.32 mM concentrations, have the same slopes and as a consequence, the solutions have the same sensitivity. There is a less complex agent in the lower concentration solution and therefore it will be not sensitive to increasing the dose.

As it can be seen from the results in Table 1, the linearity of the responses are 100 to 1000 Gy, 300 to 1000 Gy, and 100 to 500 Gy dosimetry range for the samples with a concentration of 0.32, 0.24, and 0.16 mM respectively. Except for the sample with a concentration of 0.32 mM, the dosimetric equations in this dosimetry range are logarithmic.

Figures 4, 5, and 6 show the absorption curve of the soluble samples made on the first day of irradiation at different concentrations.

Almost all Cr/DPC samples have peaks in the 540 nm wavelength range. As the results showed, with increasing the absorption dose, the height of the absorption peak decreases. This phenomenon is actually used for dosimetry. Chromium ions react with DPC to form a color complex. Because of gamma-ray irradiation and solvent radiolysis, hydrated electrons and free radicals are produced in the solvent. In the presence of H_3O + ions, chromium and DPC ions are separated and the complex color is lost due to complex fracture. Also, electrons dissolved in water and hydrogen atoms, which are highly reducers, reduce chromium ions to zero-valence chromium and prevent the returning of the reaction from re-forming the complex, and therefore the adsorption rate decreases with increasing dose. Therefore, in general, it can be concluded that despite the higher sensitivity of the samples made with lower concentrations, the range of linearity of the dosimetric response in these samples is less than in the samples with higher concentrations. Therefore, to increase the range of dosimetry response, it is better to select samples with higher concentrations.

3.2 Investigation of stability curve of Cr/DPC Chromium Nitrate dosimeter response

Stability changes over time for irradiated samples at the dose of 500 Gy can be seen in Fig. 7.

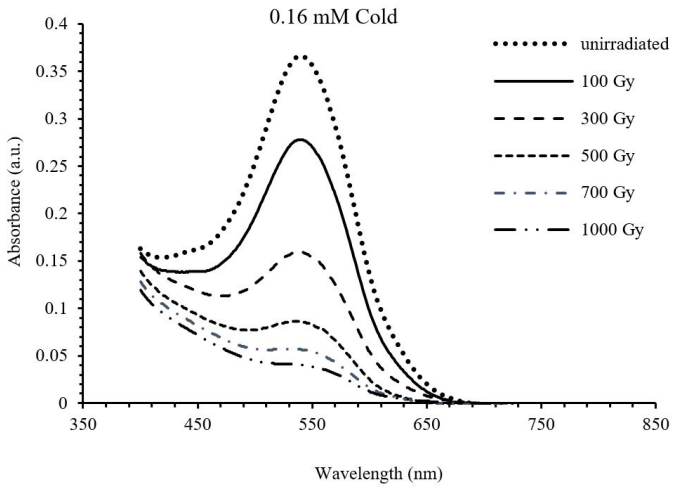

Figure 4: Absorption curve of samples made on the first day of irradiation for a concentration of 0.16 mmol.L^{-1}.

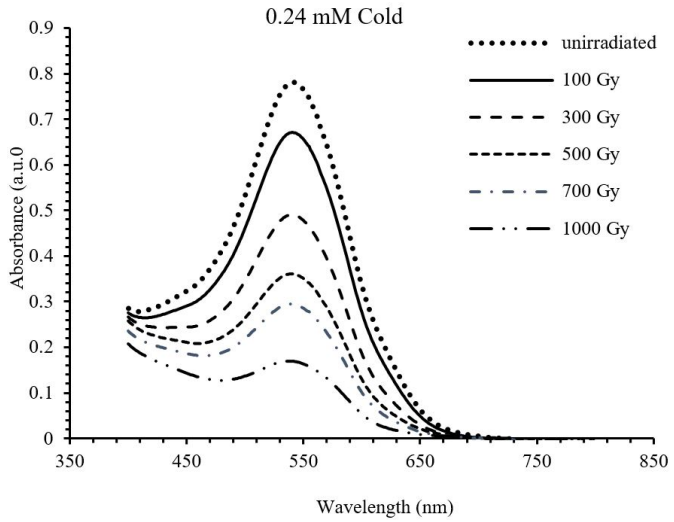

Figure 5: Absorption curve of samples made on the first day of irradiation for a concentration of 0.24 mmol.L^{-1}.

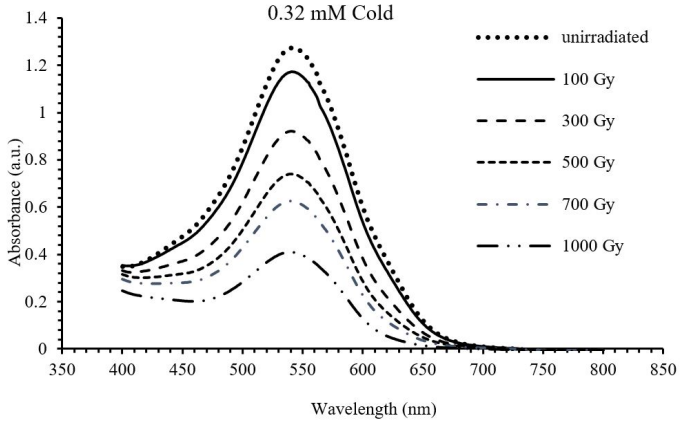

Figure 6: Absorption curve of samples made on the first day of irradiation for a concentration of 0.32 mmol.L^{-1}.

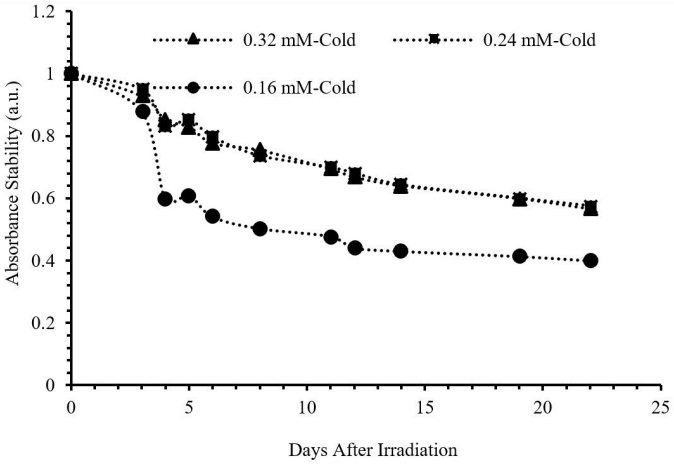

Figure 7: Stability curve of Cr/DPC irradiated samples at 500 Gy dose with different concentrations.

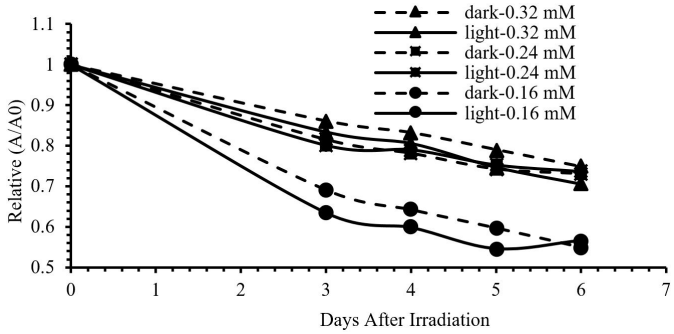

Figure 8: Stability curve of samples under different lighting conditions at dose of 500 Gy irradiation.

The results show that samples have no regular stability curves over time and even at the end of the third week, it moves towards instability. The samples with concentrations of 0.24 and 0.32 mM have 60% of their initial response after approximately 22 days after irradiation. Considering the similarity of the stability rate of 0.32 mM and 0.24 mM concentration samples, it can be concluded that increasing the concentration dose nof affect on increasing stability.

3.3 Brightness study on Cr/DPC Chromium Nitrate dosimeter response

To examine the changes in absorbance spectrum relative to the changes in light, experiments were performed in bright and dark environments. For testing in the darkness, the aluminum foil is wrapped around the test tubes and kept away from the light. Absorbance changes and relative differences of these values are presented in Table 2.

As shown in Fig. 8 and Table 2, samples that were in a dark environment had a higher absorbance height. The relative differences of absorbance for 0.32, 0.24, and 0.16 mM concentrations are approximately 13, 12, and 28%, respectively and relative changes in absorbance are greater for lower concentration samples. Figure 8 shows the stability of the samples, ie changes in absorbance compared

Table 2: Comparison of absorbance values of Chromium Nitrate solution samples in different lighting conditions.

Chromium Nitrate concentration (mM)	Absorbance (in lighting)	Absorbance (in darkness)	Percentage of relative difference
0.32	1.39	1.58	13
0.24	0.59	0.66	12
0.16	0.20	0.26	28

to the first day in different lighting conditions over 6 days. The results show that samples that were placed in a dark environment are in a more favorable condition in terms of stability than samples that were in the environment and available to light.

4 Conclusion

In this study, Chromium Nitrate solutions with concentrations of 0.16, 0.24, and 0.32 mM have been prepared and irradiated. Most chemical dosimeters do have not a wide dosimetry range. As the results show, the sample with a concentration of 0.32 mM is linear in the wide range of dosimetry 100 to 1000 Gy and the optimal concentration must be found to achieve a stable sample in about 3 weeks duration. Also, brightness study showed that the relative differences of absorbance in darkness and lighting conditions for 0.32, 0.24, and 0.16 mM concentrations are approximately 13, 12, and 28%, samples that are in a darker environment are more stable than samples that are in a lighter environment. This research can be a clear result for use in industrial applications.

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