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Spectrophotometric evaluation of a nickel-based complex solution for use as a radiation dosimeter

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HIGHLIGHTS

- A Ni/Mo solution was investigated as routine dosimeter for use in radiation processing.
- The dose response of Ni/Mo solutions was linear in the range of 50 to 1500 Gy.
- This solution was proposed as a simple construction and low cost routine dosimeter.

ABSTRACT

In this paper, the spectrophotometric properties of a colored Nickel-based solution complex (Nickel nitrate hexahydrate and Methyl Orange (MO)) were investigated as a stable chemical dosimeter with a simple synthesis and low cost for use in radiation processing. The maximum absorbance for the solution was observed at 460 nm. This solution was irradiated at three different concentrations of $Ni(No_3)_2.6H_2O$ and MO by Co-60 gamma-ray. The results showed the solution absorbance decreases with an increase in doses, and this solution can be used as a routine dosimeter and has a linear response in the 50 to 1500 Gy range with acceptable repeatability and stability in environmental conditions up to 40 days before and after irradiation.

KEYWORDS
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Spectrophotometry
Nickel complex
Methyl Orange
Gamma irradiation

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1 Introduction

Chemical dosimeters are one of the most widely used dosimetry systems. Radiochromic solutions are a type of chemical dosimeter that work based on the color change in solution due to radiation. This color change can be read by changing the optical absorbance of the solution using a spectrophotometer (Spinks and Woods, 1990). Several studies have been done on different solutes and solvents, dosimetry ranges, and different dosimetric properties on radiochromic dosimeters (Abdel-Fattah et al., 2017; Day and Stein, 1957; McLaughlin et al., 1988). Many dosimeters exist in liquid and are applied in high-dose-dosimetry applications and gels are used for the lower dose ranges (Gafar and El-Ahdal, 2016; Aldweri et al., 2017).

In 2014, Aqueous solutions of the organic dye Rhodamine B were found to be useful in measuring radiation doses in the range of 0.1 to 2 kGy and the absorption peak was observed at a wavelength of 544 nm. the absorbance readings are stable for at least 60 days in the dark at room temperature (Beshir et al., 2014).

Nitro blue tetrazolium (NBT) solution dosimeters were prepared and investigated based on radiation-induced reduction of NBT. Dosimetric properties for these dosimeters were investigated. The absorbance was increased with the absorbed dose in the dose range of 5 to 30 Gy. The stability of solution dosimeters after irradiation was very high up to 30 days. The effects of pH, irradiation temperature, and additives were investigated in this work (Rabaeh et al., 2013).

In 2016, a Fuchsine acid cyanide (FAC) dye dosimeter was introduced by Gafar and El-Ahdal (Gafar and El-Ahdal, 2016) as a stable dosimeter for the 1 to 170 Gy dose range. The dosimeter formed a colorless sample, which developed a color with an increased absorbed dose. Upon irradiation, the sample turned pink, and the

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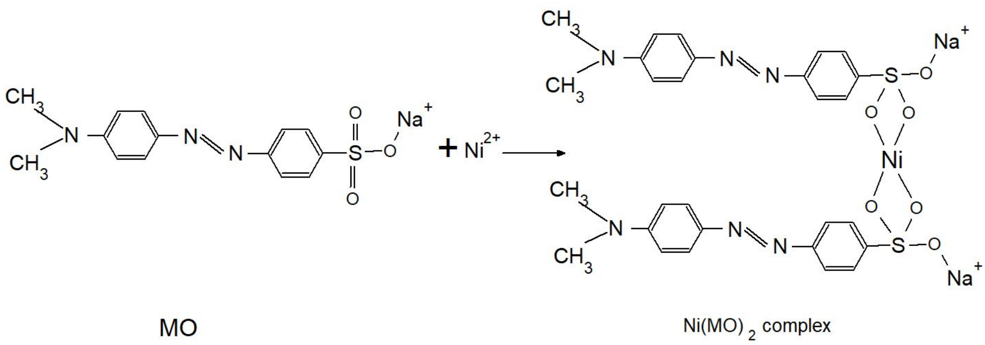

Figure 1: Chemical structure of $Ni(MO)_2$ complex.

color change was measured at 550 nm using UV-vis spectroscopy (Gafar and El-Ahdal, 2016). In 2020, a new solution dosimeter based on dithizone was introduced that showed absorbance peaks at a wavelength of 421 nm and 515 nm (Rabaeh and Basfar, 2020). The response of this dosimeter was shown to be linear up to 6 kGy. Also, the absorbance for both peaks increased gradually with increasing irradiation temperature. The absorbance of unirradiated and irradiated dithizone solution had a standard deviation of less than 3% over five days (Rabaeh and Basfar, 2020).

Nickel is a transition metal that has an incomplete substrate *d*. Due to the relatively high single-electron orbits at the *d*-level and the possibility of overlapping of these orbits and the formation of covalent bonds in the crystal lattice, these elements tend to form complex color structures (Adachi et al., 2004). A coordinate complex consists of an atom or a central ion, usually, a metal called the coordination center, and the molecules or bond ions around it are known as ligands or complex agents (Lawrance, 2013).

The purpose of this paper is to fabricate a soluble chemical metal complex dosimeters based on nickel nitrate compounds for use in irradiation of agricultural products, wastewater treatment, and food irradiation processing using gamma rays. This dosimeter is in the group of radiochromic chemical dosimeters.

Recently, a complex solution dosimeter based on Nickel nitrate and 1,5-diphenylcarbazone was investigated to measure irradiation doses in the 20 to 1000 Gy range with a linear response. Good repeatability, simple construction, low cost, and minimum detectable dose of 20 Gy, make this dosimeter a suitable option for use in radiation processing of agricultural products (Babaei et al., 2022). In this paper, were trying to increase the dose range, stability, and repeatability of the nickel-based complex solution at previous work (Babaei et al., 2022) by changing the ligand and optimizing the pH of the nickel solution complex.

2 Materials and Methods

A 0.014 g of $Ni(No_3)_2.6H_2O$ (Mw 290.7 g/mol, MERCK, Germany), and 0.016 g of Methyl Orange (Mw 327.3 $g.mol^{-1}$, MERCK, Germany) were dissolved in 50 mL ethanol and distilled water respectively, to prepare 1 mM stack solutions. The solutions were stirred separately at room temperature for 3 h to ensure the homogenous dye stock solutions. The appropriate volume of stock solutions was mixed and diluted with distilled water and ethanol solvent (volume ratio of 1:1) to obtain concentrations of 0.02, 0.03, and 0.06 mM of Ni and MO. The volume ratio was obtained by experiments to achieve maximum absorbance and stability. Figure 1 shows the chemical structure of the Ni/MO liquid complex (Siddalingaiah and Naik, 2001).

Samples were irradiated with a GC-220 irradiator at a dose rate of 1.17 $Gy.s^{-1}$. This irradiator was calibrated using ferrous sulfate (Fricke) reference dosimeter (ASTM, 2013). The samples were poured into $10 \times 10 \times 40$ mm^3 quartz cuvettes (optical path length of 10 mm), and the absorption spectra of the solutions were measured in the 400 to 800 nm wavelength range, by BECKMAN Coulter-Du 800 UV-Vis spectrophotometer. The pH of the samples was measured by METTLER TOLEDO (ASCC-14180) digital pH meter.

3 Results and discussion

Hydrochloric acid (HCl) and sodium hydroxide (NaOH) solution (0.1 M) were used to adjust the pH of the solution in all samples. Figure 2 shows the effect of pH on the absorbance at $0.06 + 0.06$ mM concentration for example. To achieve the best absorbance and stability at three concentrations, the pH of 5.1 is optimal. At higher pHs Sedimentation was observed.

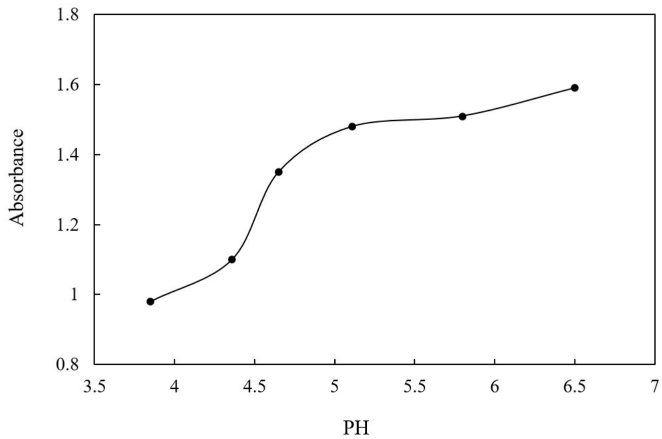

Figure 2: Effect of pH on the absorbance of the Ni/MO solution at $0.06 + 0.06$ mM concentration [Ni+MO].

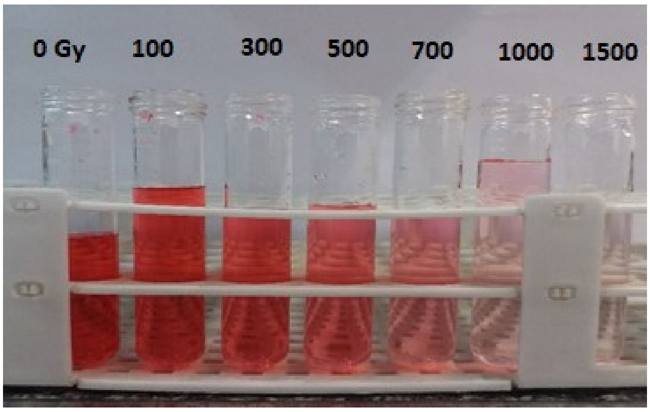

Figure 3: Color changes of Ni/MO complex solution at various Doses for $0.06 + 0.06$ mM concentration [Ni+MO].

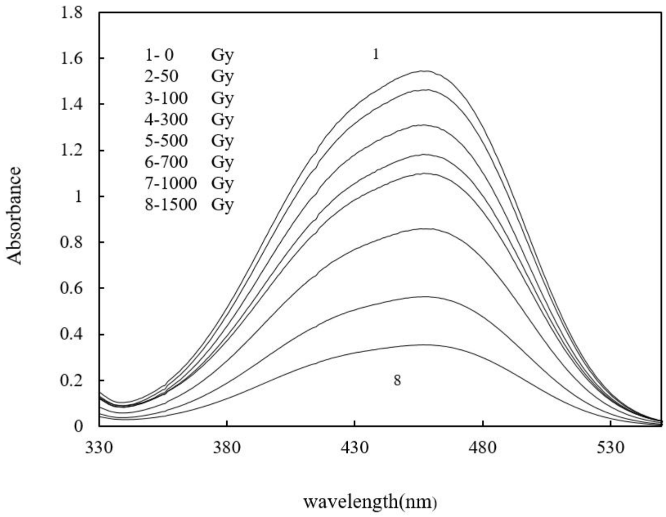

Figure 4: The absorption spectra of Ni/ MO complex solution irradiated to different absorbed doses, [$0.06 + 0.0.06$ mM].

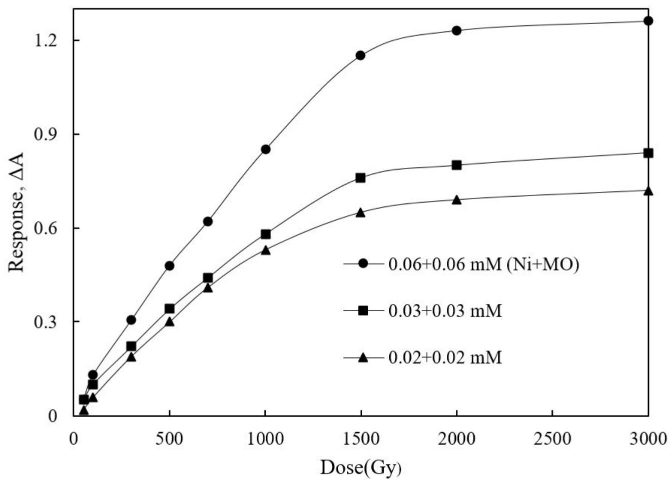

Figure 5: The dose-response curves of the Ni/ MO complex solution at 460 nm for different concentrations: $\Delta A = A0 - Ai$ where $A0$ and Ai are the optical absorptions at 460 nm wavelength for the unirradiated and irradiated solution, respectively.

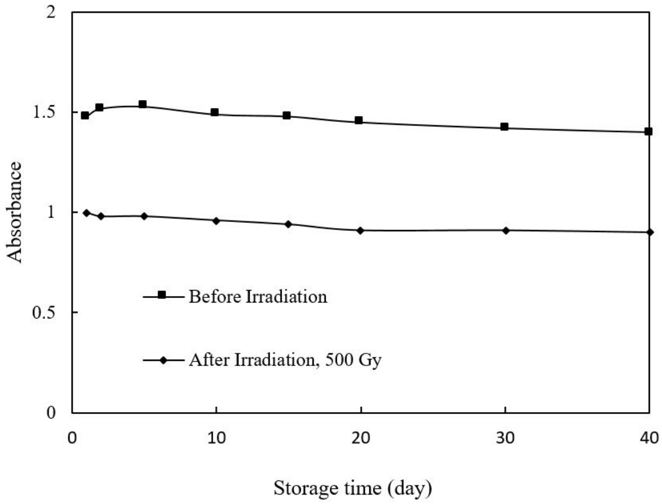

Figure 6: Stability of the liquid Ni/MO complex before and after irradiation as a function of storage time (stored in environmental conditions for $0.06 + 0.06$ mM [Ni+MO] concentration).

As shown in Fig. 3, the effect of gamma irradiation on the Ni/MO solution dosimeter is bleaching the solution color. The absorption spectra of 0.06 mM Ni and 0.06 mM MO for an un-irradiated and irradiated solution are shown in Fig. 4. The main absorbance band peaking shows at 460 nm, and absorbance decreases with an increase in doses. For the other two concentrations, the maximum absorbance peak and the behavior of the absorbance spectra are similar. Figure 5 shows the dose-response in terms of the change in the absorption peak measured at a wavelength of 460 nm versus the absorbed dose for the dose ranges of 50 to 3000 Gy for three concentrations. The linear area of the response curve is in the range of 50 to 1500 Gy with $R^2 = 0.99$.

Figure 6 shows the stability of the dosimeter ($0.06 + 0.06$ mM) before and after irradiation in environmental conditions. In fig.6, the absorbance at 460 nm has acceptable stability up to 40 days before and after irradiation. The stability for the other two concentrations is the same, which is not shown here due to similarity.

The repeatability of the dosimeters was investigated by irradiating a set of five samples for each absorbed dose and determined by the overall coefficient of variation, $CV\%$, that was calculated for the spectrophotometric measurements of the samples, by (ASTM, 2004):

$$CV(\%) = \sqrt{\frac{\sum(n_i - 1)\left(\frac{SD_i^2}{\bar{x}_i^2}\right)}{\sum(n_i - 1)}} \times 100 \qquad (1)$$

where n_i is the number of measurements of each absorbance, SD_i is the sample standard deviation, and x_i is the mean value of absorbance. The overall coefficient of variation ($CV\%$) at a confidence level of 95% over 150 data samples was determined to be 3.95%.

4 Conclusions

A Ni/MO solution was made and irradiated with a Co-60 source to investigate its spectrophotometric properties for use as a routine dosimeter for use in food and agricultural processing. The results showed that by increasing the absorbed dose the complex dissociate to Ni and MO, the optical absorption of samples was decreased and causing discoloration of solution dosimeters. As a result of optimizing the pH value (5.1) and changing the ligand (DPCO to MO), the ionic strength and the structure of the complex changes, so the linear response and stability of the Ni/Mo dosimeter increase to 50 to 1500 Gy and 40 days respectively relative to Ni/DPCO dosimeter. The repeatability of Ni/MO dosimeter is 3.95, which is better than Ni/DPCO (4.12). Finally, this solution was proposed as a simple construction and low-cost routine dosimeter which has better behaved than the previous ones at some dosimetric properties.

Conflict of Interest

The authors declare no potential conflict of interest regarding the publication of this work.

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