

STUDY OF THE METROLOGICAL CHARACTERISTICS OF THE FBX DOSIMETER IN THE PHOTON BEAM USING A SECONDARY STANDARD

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Abstract

The metrological characteristics of the dosimetric system containing 0.20 mM ferrous ammonium sulphate, 5.0 mM benzoic acid and 0.20mM xylenol orange in 0.05 N sulphuric acid. (FBX dosimeter) was investigated. The wavelength and absorbance linearity calibration of the spectrophotometer were checked using NBS Standard Reference Material. The molar absorption coefficient ϵ of the dosimeter solution was determined using carefully prepared standard solution. The G-value for the ferric-xylenol orange complex when this dosimeter is exposed in air to gamma radiation was determined using a secondary standard (ionization chamber). The dosimetric solutions could be stored for about 2 weeks before irradiations and up to 2 days after irradiations without any significant error in dose estimations. The linearity of the absorbed dose with the increases in absorbance of the dosimeter solution has been checked. For this purpose, the dosimeter solutions were irradiated to a series of different absorbed doses (3 to 11 Gy). The quality data, as judged from the correlation coefficient, demonstrate that the curve is linear in the range investigated. The stability and reproducibility of response are such that this system should be used to measure the low doses. The reproducibility allowed us to determine the lower detection limit of the FBX dosimeter, which is around 5 Gy.

Keys words: *FBX Dosimeter, Metrological, Spectrophotometer.*

INTRODUCTION

Radiation dosimetry deals with the determination of absorbed dose to the medium exposed to ionizing radiation. Liquid chemical dosimeters are very useful for the measurement of energy absorbed from ionizing radiation since the liquid can fill any shape to measure the average absorbed energy in the volume. So far, the Fricke dosimeter, also called the ferrous sulfate dosimeter, is one of the most useful chemical dosimeter that could be used with accuracy. It is useable in the dose range of 40-400 Gy. The characteristic parameters of the Fricke dosimeter made the object of an evaluation the works of which were published by diverse authors [1,2]. However, after bibliographical study, it emerges that the value of the

radiochemical yield $G(\text{Fe}^{3+})$ was determined by comparison to values of dose measured by means of a calorimeter or of a primary standard ionization chamber [3,4]. As part of a study related to chemical dosimetry, we have investigated and characterized the Fricke dosimeter in the Secondary Standard Dosimetry Laboratory (SSDL) of the Center of Radioprotection and safety (Algiers) by using a secondary standard such as a cylindrical ionization chamber calibrated in terms of air kerma. in a standard laboratory's reference quality beam, generally taken as a ^{60}Co gamma ray beam, the obtained results have been published [5]. During this work it was found that the Fricke dosimeter exhibited several attractive qualities but its application for low dose measurements has been limited by low sensitivity which is about 20 Gy for the Fricke dosimeter developed at our laboratory. Hence it was decided to investigate the system to improve the sensitivity to be able to use it in clinical beam. For this purpose several techniques were investigated by several authors. Among these techniques, The FBX dosimetry system which consists of 0.20 mM of ferrous ammonium sulphate, 5.0 mM of benzoic acid, 0.20 mM of xylenol orange in 0.05 N sulphuric acid, developed for low level dose measurements by Gupta and co-workers as a modification of the Fricke system [6,7]. Since this dosimetry system is being used as a clinical dosimeter at present, we suggested developing it in our laboratory. However, certain modifications were carried out in the method of the preparation, storage of the dosimetry solution of the FBX dosimeter. This work reports detailed investigations regarding the $G(\text{Fe}^{3+})$ determination using a secondary standard quoted above, the molar absorption coefficient using xylenol orange from Fluka manufacturer and study of the dose response, the stability and reproducibility of the FBX dosimeter. The values measured were compared to the published values. It seems that xylenol orange contains the impurities which affect the molar absorption coefficient value and the G value.

MATERIAL AND METHOD

Irradiation

All irradiations in this experiment were performed in ^{60}Co gamma rays using an Eldorado 78 unit. The output in terms of air kerma was 1250 mGy/min, at a source-to-detector-distance (SDD) of 85 cm, and a field size of $10 \times 10 \text{ cm}^2$ at the chamber axis Figure 1. The beam flatness at these conditions is about 0.3% within ± 4 cm of the beam axis. Dose estimation was done with the secondary standard ionisation chamber WDIC70 #141 calibrated at the IAEA's reference laboratory. The calibration coefficient N_k , established at the conditions $T = 20 \text{ }^\circ\text{C}$ and $P = 101.325 \text{ kPa}$, is thus traceable to the Bureau International des Poids et Mesures (International Bureau of Weights and Measures).

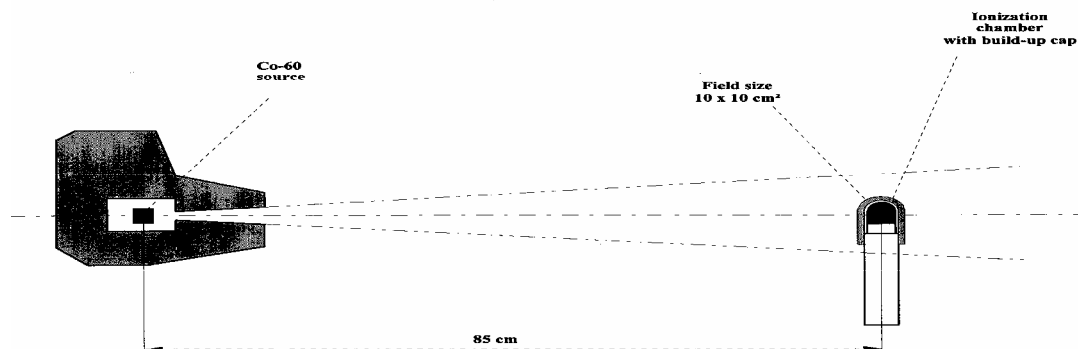


Figure 1. The geometrical conditions used for calibration in terms of air kerma at the Algerian SSDL

CHEMICAL DOSIMETER

The FBX dosimeter was prepared as one stock solution from analytical grade reagents and ultra-pure water. The weight of 610.4 mg benzoic acid was added to the glass flask containing 1.4 cm³ sulfuric acid made up to 250 ml with ultra-pure water and dissolved by warming over a water bath, then, 78.4 mg ferrous ammonium sulphate and 152 mg xylenol orange were added to the glass flask together with 750 ml of ultra-pure water for a final volume of 1000 ml of the dosimetric solution. It was then stored for one day before used to stabilize the solution.

Pyrex ampoules of about 10 mm outer diameter, 0.5 mm wall thickness, 25 mm height, and capacity 2 ml filled with the dosimetry solution were used for this study. For dose measurements from cobalt-60 unit in air, a Perspex build-up cap of 2 mm thickness was provided.

For cleaning, the glassware and irradiation ampoules were filled with Freon in the liquid state (Trichlorotrifluoroethane) and kept for 24 h and then rinsed with tap water and ultra-pure water. They were then baked at 500 °C for one hour and stored. Before use, they were rinsed with fresh dosimetric solution and filled again.

The optical density of the irradiated solution was measured on a single beam spectrophotometer against the unirradiated solution. The spectrophotometer used was a DU-58 (manufactured by Beckman Instrument) which is digital and uses square-type cuvette in quartz which had an internal diameter of 10 mm. to measure volumes of 1ml. The wavelength scale of the spectrophotometer was checked with a holmium oxide filter and the linearity of the absorbance scale was checked with dichromate solution. This instrument was found very sensitive and reproducible. Although the instrument is calibrated to read a minimum of 0.005 absorbance, it will register a still lower value. Corrections were made for the appropriate blanks, kept in similar ampoules to those used for the irradiations. The difference between the absorbance as read by the irradiated solution and the blank gives the correct dose. The molar absorption coefficient of the ferric-xylenol orange complex is measured by preparing a solution containing 0.20 mM xylenol orange in 0.05 N sulphuric acid and different concentrations of ferric ions. This complex has maximum absorption at 540 nm as shown in figure 2.

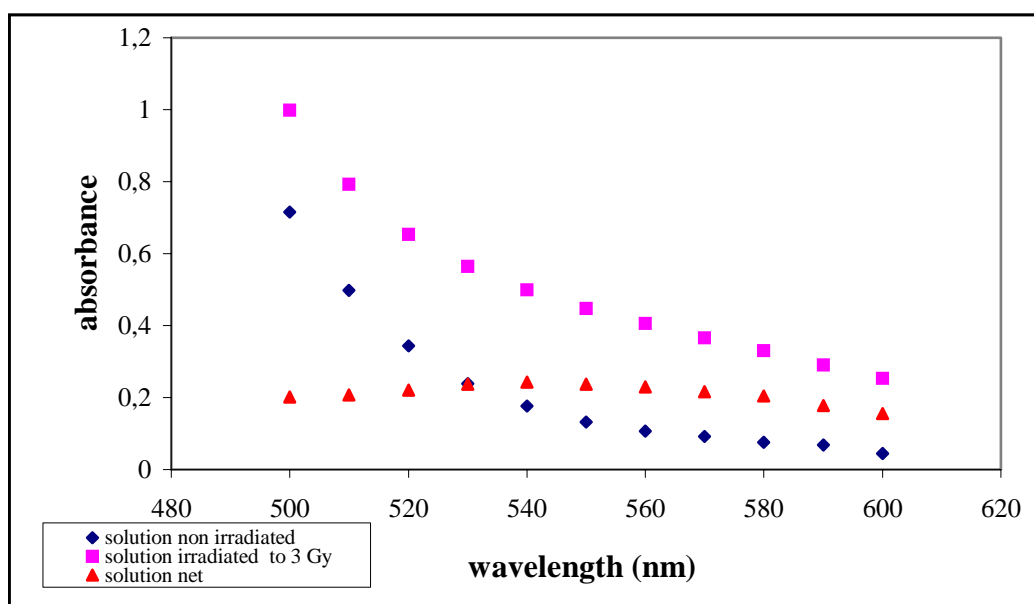


Figure 2. Absorption spectrums of the ferric-xylene orange complex.

Table 1. Molar absorption coefficient values published In the literature [8].

Dye manufacturer	Molar absorption coefficient $M^{-1} \cdot cm^{-1}$	References
K et K laboratories	14134 ± 280	[8]
Fluka	17644 ± 383	“
Koch-light laboratories	12500 ± 225	“
Loba chimie	19200 ± 294	“
BDH	13400 ± 250	Gupta (1973b)
Hopkin and williams	13425 ± 320	[9]
Merk	14207 ± 320	Pejuan (1981)
BDH	14290 ± 220	Maughan (1983)
Fluka	12175 ± 61	Present work

Absorbance versus ferric sulphate ammonium concentration and molar extinction coefficient determination

The ferric-xylene orange complex solution was prepared from analytical grade reagents and ultra-pure water. Weight of 78.4 mg of ferric ammonium sulphate, 152 mg of xylene orange, 1.4 cm³ of concentrated sulphuric acid, were added to the glass flask together with ultra-pure water for a final volume of 1000 ml of the complex solution. To find the connection between the ferric sulphate ammonium concentration and the absorbance a series of samples was prepared by diluting the prepared solution with a range of concentrations from $2 \cdot 10^{-5}$ M to $2 \cdot 10^{-4}$ M. The Absorbance readings were done at the wavelength of 540 nm for each concentration. The molar absorption coefficient ϵ was obtained from the slope of the plot of optical density against ferric ion concentration as shown in figure 3. The ϵ -value is found to be $12175 \pm 61 M^{-1} \cdot cm^{-1}$. The error given is the relative mean deviation of the four measurements. In order to validate the experimental techniques adopted by us, we have done the comparison of the ϵ -value thus determined with the published ϵ -value summarized in table 1.

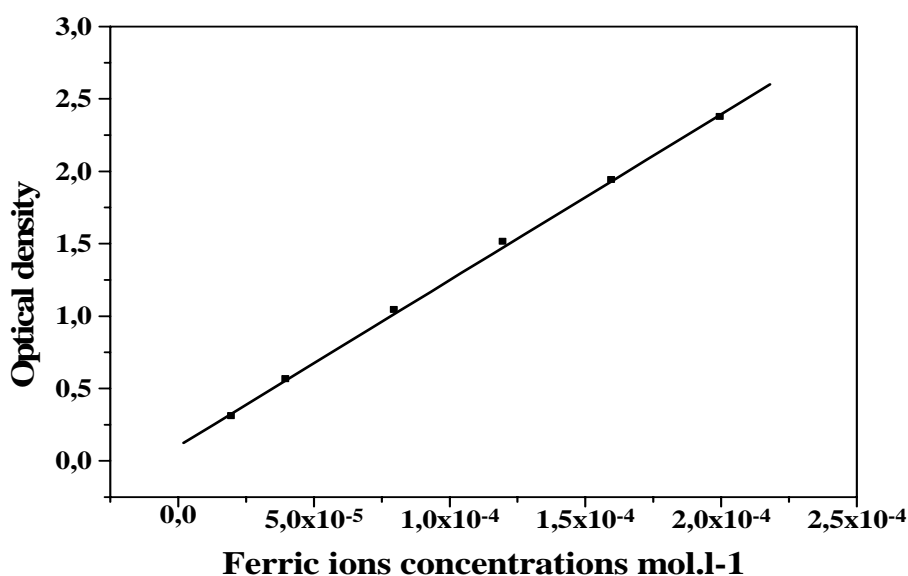


Figure 3. Absorption of Ferric ammonium sulphate in 0.05 N H₂SO₄.

Table 2. Reproducibility check carried out with FBX dosimeter.

Dose (cGy)	I	II	III	Ave	SD
300	0.321	0.315	0.322	0.319	0.0037.
500	0.422	0.417	0.418	0.418	0.0036
700	0.571	0.578	0.569	0.572	0.0047
900	0.717	0.714	0.712	0.714	0.0025
1100	0.866	0.861	0.869	0.865	0.0040

DOSE RESPONSE

The dose response of the prepared dosimeter was studied by irradiating the dosimeter with a range of doses from 300 cGy to 1100 cGy at 85 cm in air for ⁶⁰Co photons beam. The procedure was repeated thrice to check the reproducibility of the dosimeter. The readings obtained are tabulated in table 2. The dose response of the dosimeter was verified by plotting the measured optical density against the exposed dose as shown in figure 4.

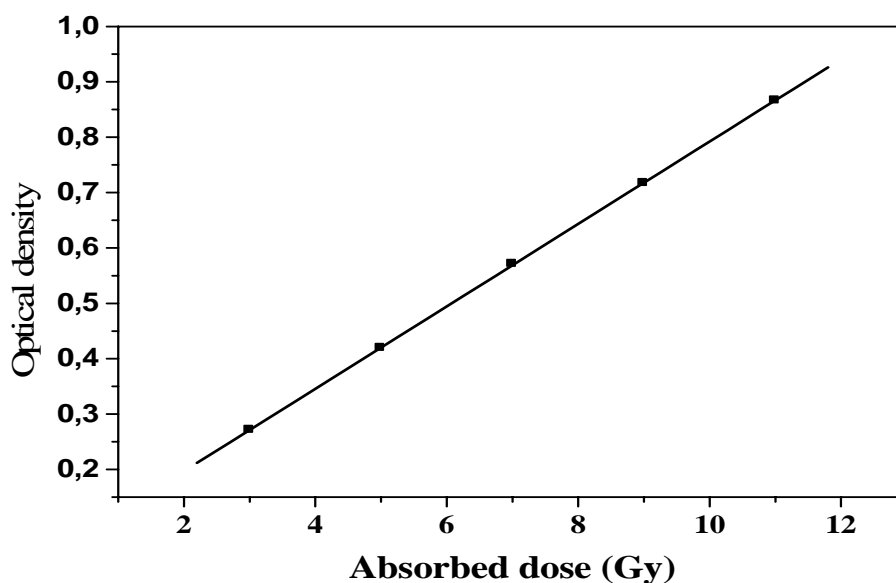


Figure 4. Calibration curve for the FBX dosimeter for Co-60 photons.

Table 3. Stability for the FBX dosimeter stored and irradiated to a dose of 11Gy in Pyrex glass flasks.

Pre-irradiation Storage (weeks)	absorbance at 540 nm for post irradiation storage (weeks)			
	0	1	2	3
1	0.866	0.842	0.825	0.815
2	0.862	0.839	0.820	0.812
3	0.858	0.845	0.827	0.813

POST-IRRADIATION STABILITY STUDY

In order to study the post irradiation stability of the absorbance changes, the dosimeter solution was stored in Pyrex glass flasks up to three weeks before irradiation and exposed to a dose of 11Gy thereafter in the glass flask itself. Then the time profiles of the absorbencies at 540 nm were measured using unirradiated solution as a blank for spectrophotometric measurements. Between each reading the dosimeter solution was stored in the dark at room temperature. The results are summarised in table 3.

RESULTS AND DISCUSSION

Table 1 represents a comparison of the molar absorption coefficient values for the dyes obtained from different manufacturers, measured by other laboratories. From this table it is quite clear that different samples of the dye still contain some impurities which affect the complex formation [8]. The dye obtained from Loba Chemie gives the highest value for the complex. Since the impurities affect the molar absorption coefficient of the ferric-xylene orange complex in the FBX system, so the largest variation between the ϵ -value determined

for our set-up and the value reported by Gupta using the same dye obtained from Fluka is due to the presence of impurities in both the xylenol orange itself because in our case the ϵ -value was carried out with the dye supplied by fluka used as received possibly more pure while in the literature the ϵ -value was determined using the dye obtained 10-15 years ago [8]. The ultra pure water used to prepare the solution has led to this largest variation because it's certainly more pure than the distilled water used in the literature.

Table 2. shows the results of the reproducibility check carried out with the FBX dosimeter. The standard deviation is found to be less than 1% of the mean dose. The difference in readings is due may be to the inherent variation in the vials. It was concluded that the prepared FBX solution is reproducible and selecting vials with less variation can further reduce the variation in readings. The dose response of the FBX chemical dosimeter from figure 4 is found to be linear for the range of doses exposed.

Table 3.gives the results of post irradiation stability studies. It is seen from the table that the reduction in absorbance due to 3 week post-irradiation storage is less than 6 %. Unirradiated solution was used as a blank for spectrophotometric measurements. However, this change is less than 2% in a week.

We also performed $G(\text{Fe}^{3+})$ calculations for comparison with published values. The formula used for calculation is based on the Beer–Lambert law and can be written as $G(\text{mol J}^{-1}) = (\text{OD}) / (D \times \rho \times \epsilon \times l)$ where D is the dose (Gy), ρ is the density (kg/m^3) of the absorbing medium, OD is measured optical density or absorbance, l is path length (m) in the cuvette and ϵ is molar absorption coefficient of the ferric xylenol complex in $\text{mol}^{-1} \text{m}^2$. The value of the molar absorption coefficient experimentally determined for our set-up ($1217.5 \text{ mol}^{-1} \text{m}^2 \pm 0.5\%$) was taken. The value of $G(\text{Fe}^{3+})$ is found to be $66 \times 10^{-7} \text{ mol J}^{-1} (\pm 1.26\%)$. The error given is the overall uncertainty due to individual uncertainties in various terms, namely number of dose (1.04.0%), molar absorption coefficient (0.5%) and absorbance (0.45%). In terms of random and non-random components, the uncertainty values are 0.5% and 1.16%. The overall uncertainty is calculated by summing the individual uncertainties in a quadratic manner. The published G -value for cobalt-60 gamma rays is $67.3 \times 10^{-7} \text{ mol J}^{-1}$ [7]. We wish to point out that the determination of G -values and then the comparison of the G -value thus determined with the published G -value, in the case of gamma rays, was done in order to validate the experimental techniques adopted by us.

CONCLUSION

From the various checks carried out with the FBX chemical dosimeter under our standard laboratory conditions it is concluded that the FBX dosimeter can be used for low dose measurements with a good reproducibility, though the pre-irradiated solutions can be stored up 3 weeks, it is necessary to measure the irradiated solution within a day of the completion of irradiation. The reagents used in the preparation of the dosimeter solution are available and the preparation does not involve complex procedures. Since the molar absorption coefficient and the G -value of the ferric-xylenol orange complex at 540 nm may change with different makes of xylenol orange, therefore, the molar absorption coefficient and the G -value should be measured for the available xylenol orange. However, to confirm our results in terms of the G -value, we plan to carry out the experiment with expected the $G(\text{Fe}^{3+})$ values in the FBX dosimeter for high-energy photons and electrons, we also intend to investigated the effect of some parameters on the response of the FBX dosimeter.

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