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Study Of Chemical Dosimetery Using Ceric Sulfate Calibration Dosimetric Method

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Abstract

In this study triply distilled water was used to make dosimeters. Ceric sulfate dosimeter and Fricke dosimeter were prepared to calibrate the dose rate in Mark-IV irradiation chamber 22×9×26 inch. The prepared solutions were placed at different part of the Mark–IV irradiation chamber, after irradiation optical density of all radiated solutions was measured by spectrophotometer at a wavelength 304nm in UV region. The variation in dose rate of both proposed dosimeters in the chamber was analyzed. Average dose rate was measured and data was analyzed statistically. Standard curves were prepared for comparative study and standardization of the proposed dosimeters was confirmed.

Keywords: Ceric Sulfate dosimeters, Ferric sulfate dosimeters, Mark-IV irradiator, optical density, dose rate standardization

Introduction

These radiations can cause biological, chemical as well as physical changes in the matter through which they pass. Radiations cause biological changes, which result in the killing of living organisms and bacteria etc from single cell to large animals [1]. The gamma rays are electromagnetic radiations emitted from excited atomic nuclei as an integral part

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of the process whereby the nucleus rearranges itself into a state of lower excitation. The wavelength of gamma radiation which is the characteristics of the emitting substance with proper range from 8.9×10^{-10} to 4.7×10^{-13} meter. The gamma radiations emitted by radioisotopes are either mono-energetic or have a small number of discrete energies as for example Co^{60} which have energies 1.33 MeV and 1.17 MeV. The mean energy of Co^{60} is 1.25 MeV. Ionizing radiations when interact with biomass can damage the living cells because of which the cells either die or change their structure and function. If damage is minor naturally the body repairs itself easily after a certain interval of time but heavy damages are irreparable. The high intensity ionizing radiations excites the atoms and molecules, and thereby promoting chemical reactions on interactions with matter. Gamma rays are highly penetrating and can, therefore, reach easily to the internal organs of the body.

Chemical dosimetry is still an active research area of research in the present era. Using cobalt 60 becomes and active areas of research

The radioactive isotope obtained when cobalt is subjected to a neutron bombardment in the nuclear pile.

Following is the decay scheme;

$$Co^{60} \longrightarrow Co^{60} \longrightarrow Ni^{60} \longrightarrow Ni^{60}$$

$$\gamma\ (0.59\ MeV) \qquad \beta\ (0.31MeV) \qquad \gamma(1.17MeV) \qquad \gamma(1.33MeV)$$

Each disintegration of a radioactive Cobalt nucleus therefore gives rise to two γ–photons, of energy 1.33MeV and 1.17 MeV, as well as some β and γ,s that are mainly absorbed in source. This is notably the case of Co^{60} the energy output per curie is $5.92\times(1.33+1.77)=14.8$ m watt/curie.

Source containing activities between 100 and 1000 Ci are in common use in laboratories. [2].

Chemical dosimetry is based on the determination of the radiation dose from the chemical change produced in an irradiated medium. But the sensitivity of a system to

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radiation can be expressed in terms of the changes produced by a given radiation dose. The desirable characteristics of any chemical dosimeter are Satisfactory sensitivity, Adequate re-predictability, Stability under normal conditions before and after irradiation, Independence of product yield on dose rate, quality of radiation and temperature, pH oxygen and reactive solute concentration in aqueous system, Short duration of post irradiation changes if present.

In practice no single dosimeter meets all these requirements because of the complexity of phenomena occurring when substance is irradiated. As for a chemical system Fricke dosimeter developed by Fricke and Morse, which is based on the oxidation of an aerated ferrous sulfate solution meet most of the above condition dose measurement.

Later on, Miller developed it. The Fricke dosimeter has been accepted as a standard dosimeter for the calibration of other dosimeters. In the Fricke dosimeter, when ferrous sulfate solution is irradiated with gamma radiations, the ferrous ions Fe^{++} are converted into ferric ions Fe^{+++}. For a Co^{60} gamma source, the yield of Fe^{+++} is 15.6 ions per 100 eV. This chemical change is determined by measuring the absorbance of the solution at 304 nm wavelengths by spectrophotometer, which is a direct measured of the absorbed dose. The dose range of Fricke dosimeters is from 40 to 400 Gy for 304 nm wavelengths.

The Ceric sulfate dosimeter can be used to measure doses in the range 0.01 to 100 M rad. It is the only chemical dosimeter that may be regarded as a reference standard for aerated ferrous sulfate solution. However, the most serious disadvantage of Ceric sulfate dosimeter is its extreme sensitivity to impurities. On the other hand it has the advantage that the yield is independent of the presence of oxygen. As the range of Ceric sulfate dosimeter is 10^3 to 10^8 rad at wavelength 320 nm. The range can be varied by changing the concentration of Ceric sulfate in the solution. Analytic grade chemicals and high purity water will prepare this dosimeter. The dosimeter solution will be irradiated by Mark-IV (Gamma) irradiation. This irradiated solution will be stored under the low temperature and the dose stability comparison of Fricke and Ceric sulfate dosimeters will be made by measuring the change in absorbed dose [3-8].

In this research project, Fricke and Ceric sulfate dosimeters were prepared from analytical grade chemicals and triple distilled water. These dosimeter solutions were placed at different locations in the Mark-IV irradiation chamber 22″×9″×26″ and were irradiated by Mark-IV irradiator at NIAB. For 3 hours in air media. The absorbance of all the dosimeters was measured spectrophotometrically at a wavelength of 304 nm. The

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variation in absorbance of both dosimeters with respect to their location in the chamber was studied graphically and by drawing bar charts. Average absorbance of this chamber for both dosimeters was also calculated. To calculate the dose of both dosimeters in the chambers, six samples of both dosimeters were irradiated at regular equal intervals of irradiation time. The absorbance of these sample dosimeters was measured spectrophotometrically at 304 nm wavelengths. Absorbed dose was calculated for Fricke dosimeters and curve was drawn between absorbance and absorbed dose and doses of any Fricke dosimeter in the chamber were determined from these standard curves. Similarly standard curves were drawn between absorbance of Ceric sulfate dosimeter and dose of the Fricke dosimeter and from these standard curves, doses of Ceric sulfate dosimeters in the chamber were determined.

The objectives of this study are to measure absorbed dose rate of Fricke and Ceric sulfate dosimeters in different portions for calibration of the chamber and also find the coefficient of variation (C.V)% of the dosimeters for the stability.

Material And Method

Our main concern in this research project is related to comparative study of absorbance between Fricke and Ceric sulfate dosimeter in the Mark-IV irradiation chamber. Absorbance dose was also determined by means of standard curves. Spectrophotometer (CECIL, 1021), Mark-IV irradiation (γ–rays source) assembly, Plastic bottles, Glass vials, Beakers and measuring flasks, Electronic balance, Sulfuric Acid H_2SO_4 (95-98% conc), Ferrous sulfate ($FeSO_4.7H_2O$), Sodium Chloride (NaCl), Ceric sulfate ($Ce(SO_4)_2.4H_2O$), Triply distilled water.

The standard Fricke dosimeter was prepared from analytical grade chemicals and triply distilled water. First of all chemicals were weighed by electric balance then 0.28 gm $FeSO_4.7H_2O$ and 0.06 g of NaCl were dissolved in triply distilled water separately and then mixed together in a measuring flask. Then 22 ml of H_2SO_4 was added in it and adding made one liter of final solution triply distilled in the above solution. This solution was poured into 24 glass vials and was sealed off carefully. These glass vials were marked as $1D_{11}$, $1D_{12}$, $1D_{13}$, $1D_{31}$, $1D_{32}$ ------ $4D_{32}$, $4D_{33}$.

These dosimeters were placed in four portions of the chamber in such a manner that Fricke dosimeters in were in first and third rows in each portion. There were six

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Fricke dosimeters in each portion. Mark-IV irradiator irradiated these dosimeter solutions in the chamber for 3 hours in air media. The value of the optical Density (OD) or absorbance of each Fricke dosimeter at different locations in the chamber was determined by spectrophotometer at the wavelength of 304 nm. The variation in absorbance of the dosimeters at different locations in the irradiation chamber was studied graphically. The absorbance was responsible for absorbed dose in the solution.

First of all one liter of 0.8N H_2SO_4 solution was prepared by dissolving 22.22 ml of concentrated H_2SO_4 in triply distilled water. For 250 ml of 100 mM Ceric sulfate stock solution of 10.103 gm ($Ce(SO_4)_2.4H_2O$) was dissolved in 0.8N H_2SO_4. Further 2 ml of stock solution was dissolved in 0.8N H_2SO_4 to make one liter of Dosimetric solution.

This solution was poured into 12 glass vials and was sealed off carefully. These glass vials were marked as $1D_{21}$ $1D_{22,}$ $1D_{23,}$ $2D_{21}$----------$4D_{22,}$ $4D_{33.}$

These dosimeters were placed in four portions of the chamber in such a manner that Ceric sulfate dosimeters were in second row of each portion. There were three Ceric sulfate dosimeters in each portion. Mark-IV irradiator irradiated these solutions in the chamber for 3 hours in air media. The value of the optical density (OD) or absorbance of each Ceric sulfate dosimeter at different locations in the chamber was determined by spectrophotometer at the wavelength of 304nm. The variation in absorbance of the dosimeters at different locations in the irradiation chamber was studied graphically. The absorbance was responsible for absorbed dose in the solution. Mark–IV irradiation chamber (22″×9″×26″) was divided into four portions such that the distance between each portion was 6″. And each portion was divided into three rows and three columns. The distance between each row was 4″ and between each column was 7″. The Fricke dosimeters were placed in the first and third rows and Ceric sulfate dosimeters were in the second row in each portion of the chamber. All these dosimeters were irradiated simultaneously by Mark-IV irradiator for 3 hours in air media [9-14].

The value of optical density (OD) of each dosimeter was determined by spectrophotometer at a wavelength of 304nm. The absorbance (OD) of both dosimeters at different irradiation time intervals was studied graphically using regression analysis and standard curves were obtained. For Fricke dosimeter, absorbed dose was also calculated. The absorbance was responsible for absorbed dose in the solution.

DOSE CALCULATION

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The absorbed dose in "Rad" was calculated with the help of following formula.

$D_{rad} = 2.76 \times 10^4 \times \Delta OD/d$ (Rad)

Where "ΔOD" is the difference between optical density of irradiated and non-irradiated solution," d ", is the optical path length or the sample thickness.
For Fricke dosimeter, a graph between absorbed dose and time was plotted and standard curves between absorbance and absorbed dose was also drawn. Similarly for Ceric sulfate dosimeter standard curves between absorbance (OD) and absorbed dose was drawn. From these curves, with known absorbance, the absorbed doses of any dosimeter at any location in the chamber were determined [15-20].

Results

Fricke and Ceric sulfate dosimetric solutions were prepared with triply distilled water and analytical grade. Firstly, Fricke dosimeter solution was poured into 24 glass vials and was placed at different locations in the Mark-IV irradiation chamber. The chamber was divided into four portions and these dosimeters were irradiated by Mark-IV irradiator at NIAB for three hour.

The results of comparative study of deferent dosimeters are depicted briefly in tabular and graphical sketch form.

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Table 2kz1: Statistically arranged data for Fricke dosimeters in the different Portions of the Mark- IV irradiation chamber

Sr. No.	Portion in the chamber	Mean absorbance	Standard deviation (S.D)	Coefficient of variation (C.V)%
1.	Ist	1.2843	0.06969	5.4261
2.	2nd	1.3337	0.08492	6.1832
3.	3rd	1.4577	0.16228	11.1325
4.	4th	1.3523	0.06988	5.1677

Table 2kz2: Statistically arranged data for Ceric sulfate dosimeters in the different Portions of the Mark-IV irradiation chamber

Sr. No.	Portion in the chamber	Mean absorbance	Standard deviation (S.D)	Coefficient of variation (C.V)%

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1.	Ist	0.9096	0.04441	4.8821
2.	2nd	0.8953	0.04209	4.7021
3.	3rd	0.8730	0.04670	5. 3495
4.	4th	0.8880	0.05393	6. 0 727

Table 2kz3: Statistically arranged data for Fricke dosimeter in the different portions of the Mark- IV irradiation chamber

Sr. No.	**Irradiation time X**	**Absorbance**	**Estimated absorbance** $\bar{Y} = 1.239 + 0.00814X$	**e =Y- Y**
1.	1	1.176	1.24714	-0.07114
2.	2	1.236	1.25528	-0.01928

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3.	3	1.322	1.26342	0.05858
4.	4	1.354	1.27156	0.08244
5.	5	1.372	1.27970	0.09230
6.	6	1.145	1.28784	-0.14284

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Table 2kz4: Statistically arranged data for Ceric sulfate dosimeter in the Different Portions of the Mark- IV irradiation chamber

Sr. No.	Irradiation time X	Absorbance Y	Estimated absorbance $\hat{Y}$ =0.7876+0.00883X	e =Y- $\hat{Y}$
1.	1	0.798	0.79643	1.5×10^{-3}
2.	2	0.799	0.80526	-6.26×10^{-3}
3.	3	0.811	0.81409	-3.09×10^{-3}
4.	4	0.842	0.82292	0.01908
5.	5	0.820	0.83175	-0.01175
6.	6	0.841	0.84058	4.2×10^{-4}

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Table 2kz5: Statistically arranged data for the estimated absorbed dose of Fricke dosimeter

Sr. No.	**Irradiation time X**	**Absorbance** Y	**Estimated absorbed dose** $\hat{Y}$ **=1661.99+66.989X**	**e =Y- $\hat{Y}$**	**Estimated absorbed dose (Gray)**
1.	1	1104	1728.979	-624.979	17.28
2.	2	1656	1795.968	-139.968	17.95
3.	3	2447.2	1862.95	584.25	18.62
4.	4	2741.6	1929.96	811.64	19.29
5.	5	2907.6	1996.93	910.27	19.96
6.	6	823.4	2063.92	-1240.52	20.63

Table 2kz6: Statistically arranged data for estimated absorbance and estimated dose for Fricke dosimeter

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Sr. No.	Estimated absorbance	Estimated dose (Gy)
1.	0.04	17.28
2.	0.06	17.95
3.	0.08	18.62
4.	0.10	19.29
5.	0.12	19.96
6.	0.14	20.63

Table 2kz7: Statistically arranged data for estimated absorbance and estimated dose for Ceric sulfate dosimeter

Sr. No.	Estimated absorbance	Estimated dose (Gy)

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1.	0.01	17.28
2.	0.0155	17.95
3.	0.021	18.62
4.	0.027	19.29
5.	0.033	19.96
6.	0.040	20.63

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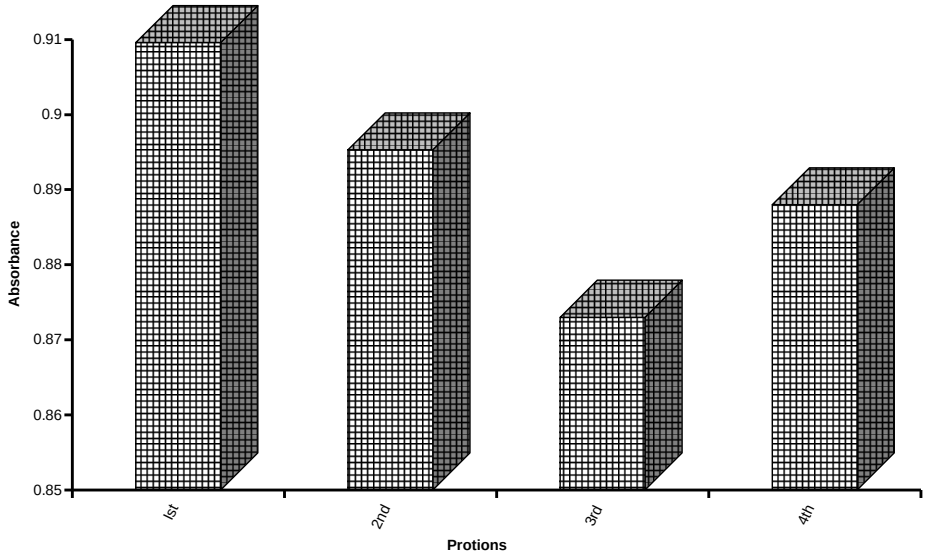
Fig 12a. Comparison of the variation in the absorbance of Ceric sulfate dosimeters between different portions of the chamber



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Discussion

Fricke and Ceric sulfate dosimeter for different portions of the chamber. Standard deviation (S.D) for both dosimeters in the each portion was calculated and recorded in the form of Table **2kz1** and **2kz2.** For the comparison between Fricke dosimeter and Ceric sulfate dosimeter in each portion of chamber, the coefficient of variation (C.V) was calculated by using the formula; C.V=S/Y×100. Coefficient of variation (C.V) was use to see that which dosimeter was not more efficient than the other dosimeter. C.V in the Ist portion for Fricke dosimeter and Ceric sulfate dosimeter was 5.4261% and 4.8821% respectively, which showed that Ceric sulfate dosimeter was more consistent than Fricke dosimeter. To see how much it was precise, the difference between their C.V taken, which was 0.544% i.e., Ceric sulfate dosimeter was 0.544% more consistent than Fricke dosimeter in the first portion of the chamber. Similarly in the second portion of the chamber, the C.V of Fricke dosimeter and Ceric sulfate dosimeter was 6.1832% and 4.7021% respectively % i.e., Ceric sulfate dosimeter was 1.4811% more consistent than Fricke dosimeter in the second portion of the chamber. The C.V of Fricke and Ceric sulfate dosimeter in third portion was 11.1325% and 5.3495% respectively; Ceric sulfate dosimeter was 5.783% more reliable than Fricke dosimeter. In the fourth portion of chamber C.V of the Fricke dosimeter and Ceric sulfate dosimeter was 5.1677% and 6.0727% respectively, which showed that Fricke dosimeter was 0.905% precise than Ceric sulfate dosimeter. Average absorbance for both dosimeters was calculated and comparison of the average absorbance and Ceric sulfate dosimeter was studied graphically as shown in Fig. **2kz1**.

It was clear that Fricke dosimeter showed maximum absorbance and Ceric sulfate dosimeter showed minimum absorbance in the Mark-IV irradiation chamber. Standard

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deviation (S.D) for both dosimeters was also calculated. To see the variability between both dosimeters, coefficient of variation was calculated by using the formula C.V=σ/Y×100 where σ is S. D and Y is mean absorbance. C.V for Fricke and Ceric sulfate dosimeter was 6.9774% and 5.2516% respectively which showed that the Ceric Sulfate dosimeter 1.7258% more reliable than Fricke dosimeter. Fricke dosimeter and Ceric sulfate dosimeter were prepared from the same procedure with triply distilled water. Each dosimeter solution was poured into six glass vials. Six samples of Fricke dosimeter were donated as A, B, C, D, E, and F, and similarly six samples of Ceric sulfate dosimeter were donated as Á, Ḃ, Ć, Ḋ, É and Ḟ. These samples were loaded in the Mark-IV irradiation chamber and were irradiated at regular equal intervals of irradiation time. The samples A and Á were irradiated for 1hour, B and Ḃ were irradiated for 2 hours, C and Ć were irradiated for 3 hours, D and Ḋ were irradiated 4 hours E and É were irradiated for 5 hours and lastly the sample F and Ḟ were irradiated for 6 hours. The absorbance of each dosimeter at regular intervals of irradiation time was measured spectrophotometrically at the wavelength of 304nm [21-25].

The data of absorbance for both dosimeters were recorded in the form of table. The data were statistically analyzed and curves were fitted. The bested fitted equation was straight-line i.e., Y=a+bX. The graph between estimated absorbance and irradiation time for both dosimeter solutions was plotted. For Fricke dosimeter, estimated absorbance and irradiation time at 304 nm wavelength is given in the Table **2kz3.** The irradiation of this dosimeter solution prepared in triply distilled water induced the conversation of ferrous ions into ferric ions. The absorbance of post irradiation solution was measure spectrophotometrically at 304 nm wavelength. The observed data was analyzed statistically and linear curves were fitted to the data. Y-intercept and regression coefficient was calculated. The Y-intercept was found to be 1.239 and regression

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coefficient was 0.00814 which showed per hour increase in absorbance with increasing irradiation time. The estimated absorbance was calculated by the equation Y=a+bX. the estimated absorbance was plotted against irradiation time at 304 nm wavelength in the Table **2kz3**.The estimate of error or residual effect was also calculated from the observed absorbance Y and estimated absorbance $\hat{Y}$. Standard deviation of regression or standard error of estimate was also calculated by the formula.

$$SE = \sqrt{\frac{\sum (Y - \hat{Y})^2}{(n - 2)}}$$ which was 0.10557348.

For Ceric sulfate dosimeter, estimated absorbance and irradiation time at 304 nm wavelength is given in Table **2kz4-2kz7.** The absorbance of post irradiation of Ceric sulfate dosimeter solution was measure spectrophotometrically at 304 nm wavelength. The observed data was analyzed statistically and linear curves were fitted to the data. Y-intercept and regression coefficient was calculated. The Y-intercept was 0.7876 and regression coefficient was 0.00883 which showed per hour increase in absorbance with increasing irradiation time. The estimated absorbance was calculated by the equation Y=a+bX. the estimated absorbance was plotted against irradiation time at 304 nm wavelength. The estimate of error or residual effect was also calculated from the observed absorbance Y and estimated absorbance $\hat{Y}$. Standard deviation of regression or standard error of estimate was also calculated by the formula.

$$Se = \sqrt{\frac{\sum (Y - \hat{Y})^2}{(n - 2)}}$$ which was 0.0117608.

The absorbed dose of Fricke “dosimeter was calculated in “ Rad” with the help of absorbance or optical density by using the formula.

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$D_{rad} = 2.76\times10^{4}\times\Delta OD/d$ (Rad)

The linear curve was fitted to the data. Intercept and slop were calculated. The regression coefficient was 1661.99. The Y- intercept was to be found to be 66.989. The estimated value of absorbed dose was also calculated with help of regression line equation Y = a + b X. the absorbed dose was plotted against irradiation time. The estimate of error "e" was calculated from observed dose and estimated dose. Standard deviation of regression was also calculated by the formula.

$$Se = \sqrt{\frac{\sum (Y-\hat{Y})^2}{(n-2)}}$$ which was 971.825.

To calculate the value of absorbed dose of Fricke dosimeter at any location in the e Mark-IV irradiation chamber, standard curve was drawn between estimated absorbance and estimated absorbed dose of the Fricke dosimeter. With known absorbance of the Fricke dosimeter the value of absorbed dose in Gray (1Gy=100Rad) could be calculated by mean of standard curves by drawing perpendiculars on the curve. If the absorbance of Fricke dosimeter was 0.39 than estimated absorbed dose was 120.0 Gy. Similarly if absorbance was 0.2236 and 0.62 than the estimated absorbed dose was 68.0 Gy and 186.2 Gy respectively. Calculation of dose from standard curves for Ceric sulfate dosimeter. Similarly for Ceric sulfate dosimeter, the absorbed dose can be calculated by mean of standard curves. Standard curves for Ceric sulfate dosimeter was drawn between estimated absorbance of Ceric sulfate dosimeter and the estimated dose of Fricke dosimeter. If the absorbance of Ceric sulfate was 0.014 then the estimated absorbed dose was 62.4 Gy. Similarly if absorbance was 0.021 and 0.027 than the estimated absorbed dose was 163 Gy and 249.6 Gy respectively [26-28].

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