Radiation Dose Measurement Using Chemical Dosimeters

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1. Introduction

The radiation dose can be estimated in various ways. Dose estimates can be obtained by either experiment or theoretical analysis. In experiments, radiation impact is assessed by measuring any change caused by energy deposition to the exposed matter, in terms of energy state (physical change), chemical production (chemical change) or biological abnormality (biological change).

The chemical dosimetry is based on the implication that the energy deposited to the matter can be inferred from the consequential change in chemical production. The chemical dosimetry usually works on the sample that is an aqueous solution, a biological matter, or an organic substance [1].

In this study, we estimated absorbed doses by quantitating chemical changes in matter caused by radiation exposure. Two different chemical dosimeters, Fricke and ECB (Ethanol-Chlorobenzene) dosimeter, were compared in several features including efficacy as dose indicator and effective dose range.

2. Materials and Methods

2.1 Fricke dosimeter

The aqueous ferrous sulphate dosimeter, usually called Fricke dosimeter, operates based on the chemical process of oxidation of ferrous ions (Fe^{2+}) in aqueous sulphuric acid solution to ferric ions (Fe^{3+}) by radiation exposure [2]. The concentration of ferric ion production is directly related and, therefore, the absorbed dose to the aqueous sulphuric acid solution can be estimated by measuring the concentration of ferric ion production after irradiation. The concentration of ferric ions can be quantitated by spectrophotometry, where the absorbance of 350 nm electromagnetic (EM) wave indicates the concentration of ferric ions.

2.2 ECB dosimeter

The ECB dosimeter is a mixture of 94% ethanol (C_2H_5OH) and chlorobenzene (C_6H_5Cl) . The ECB dosimetry is founded on the fact that chlorobenzene is dissociated into $(C_6H_5)^+$ and Cl^- by radiation exposure and the production of Cl^- is proportional to the absorbed dose to the ECB solution. Free chlorides are combined with mercuric ions from mercuric nitrate $Hg(NO_3)_2$ in nitric acid (HNO_3) to precipitate as insoluble $HgCl_2$. The ECB dosimetry works by quantitating the endpoint of titration by mercuric nitrate (titrant) for free chlorides (titrand). The endpoint is indicated by diphenylcarbazone (DPC) which combines with the excess Hg^{2+} to turn into violet-red. The process of titration with $Hg(NO_3)_2$ for chloride concentration and the indication by DPC of the titration endpoint is as follows:

$$Cl^- + Hg(NO_3)_2 \rightarrow HgCl_2^- + NO_3^- + Hg^{2+}$$

$$Hg^{2+} + \text{DPC} \rightarrow \text{violet-red coloration}$$

2.3 Electron beam irradiation

Electron beam was available from different accelerators operating at 1MeV, 2.5MeV or 10MeV. Each accelerator operated at a varying tube current: 1 mA, 3 mA, 5 mA, 7 mA, and 9 mA. The Fricke and ECB dosimeter solutions of 25 mL in a glass container were exposed under the beam exit moving at different speeds of 2 m/min, 5 m/min, 7 m/min, 10 m/min and 20 m/min. After Irradiation is completed, the solutions were collected in test tubes to be analyzed. Shown in Fig. 1 is the irradiation process with an electron accelerator (EB Tech Co., Ltd.).

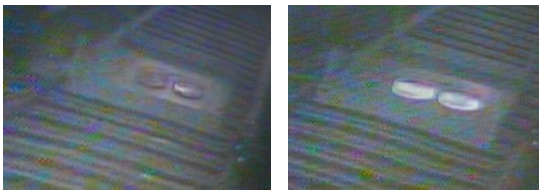

Fig. 1 Irradiation of dosimeter solutions under the beam exit of an electron accelerator.

2.4 Dose estimation with Fricke dosimeter

The Fricke dosimeter solution was prepared by mixing ferrous ammonium sulfate $(NH_4)_2Fe(SO_4)_2 \cdot 6H_2O$, sodium chloride NaCl and 95% sulfuric acid H_2SO_4. We used Fricke dosimeter solution with 10 times or 100 times of Fe^{2+} concentration given in manual. The change in EM wave absorbance is linearly proportional to the absorbed dose to the solution in some useful range. The absorbed dose to the Fricke solution is calculated by the following formula:

$$D_F = \frac{\Delta A}{\varepsilon \cdot G \cdot \rho \cdot d}$$

D_F = absorbed dose (Gy) to the Fricke solution
ΔA = net absorbance at 350 nm EM wave
ε = molar linear absorption coefficient of the ferric ions = 219 m^2/mol at 25°C.
G = radiation chemical yield of ferric ions = 1.61 x 10^{-6} mol/J at 25°C.
ρ = density of the Fricke solution
d = optical pathlength (m) of solution in the cuvette

2.5 Dose estimation with ECB dosimeter

The calibration curve in terms of the mercuric nitrate consumption (mL) versus titration endpoint or the concentration of sodium chloride in standard solution was obtained with 0.005, 0.010, 0.015 and 0.020 M of NaCl solutions. The concentration of chloride ions formed by radiolysis is calculated by

$$[Cl^-] = [Hg^{2+}] \cdot \frac{\text{(vol. of titrant) - (vol. of blank solution)}}{\text{volume of ECB solution}}$$

The absorbed dose to the ECB solution D_{ECB} is thus obtained by

$$D_{ECB} = \frac{[Cl^-]}{G(Cl^-) \cdot \rho}$$

where ρ = density of the ECB solution.

3. Results

3.1 Fricke dose estimates

The absorbed dose to water medium was obtained as in Fig. 2 by Fricke dosimetry. Dose increases with the beam current. The faster movement of glass container under the beam exit led to less radiation exposure of the Fricke solution and thus lower dose. The saturation in 350 nm EM absorbance is observed at dose levels above 600 Gy.

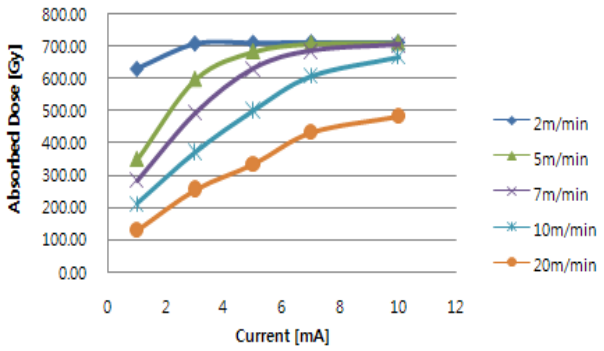

Fig. 2 Absorbed doses estimated from the Fricke solution exposed to the beam of 2.5 MeV accelerator operating at a varying beam current.

3.2 ECB dose estimates

Shown in Fig. 3 are dose estimates by ECB dosimetry with 10% and 40% ECB solutions. Dose estimates with 10% and 40% ECB solutions are comparable at low doses, which implies that the colorization by DPC is not saturated yet in 10% ECB solution. At higher doses, however, the difference in dose estimate is not negligible. The higher dose estimated with 40% ECB solution implies that the DPC colorization with 10% ECB solution is already saturated.

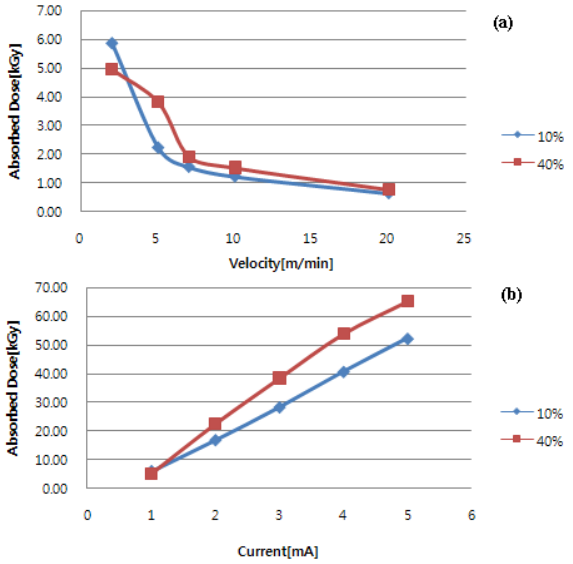

Fig. 3 Absorbed doses estimated with 10% and 40% ECB solutions exposed to the beam of 1 MeV accelerator: (a) 1 mA in beam current, (b) 2 m/min in glass container movement.

4. Conclusion

The Fricke and ECB dosimeters can be useful for measuring absorbed dose to aqueous solutions of organic or biological matters which are sensitive to UV or impurities. In chemical dosimeters, there possibly is a saturation point beyond which the dose increase does not lead to the increase in chemical index.

In this study, we have found that the Fricke dosimeter is effective at doses below 600 Gy. The ECB dosimeter was, on the other hand, saturated at doses over 6 kGy with 10% ECB solution. We are not certain yet whether the DPC colorization is not saturated with 40% ECB solution without comparing dose readings from an ECB solution of a higher CB concentration with those from 40% ECB solution.

References

[1] RJ Woods and AK Pikaev, Applied Radiation Chemistry: Radiation Processing, John Wiley & Sons, Inc., pp. 1~125 (1994)
[2] IAEA, Dosimetry for Food Irradiation, Technical Reports Series No. 409 (2002)