OXALIC ACID AS A LIQUID DOSIMETER FOR ABSORBED DOSE MEASUREMENT IN LARGE-SCALE OF SAMPLE SOLUTION

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Abstract

This study shows the feasibility for applying 2.5 mM aqueous oxalic acid solution using spectrophotometric analysis method for absorbed dose measurement from 1 to 10 kGy in a large-scale of sample solution. The optimum wavelength of 220 nm was selected. The stability of the response of the dosimeter over 25 days was better than 1 % for unirradiated and ±2% for irradiated solution. The reproducibility in the same batch was within 1%. The variation of the dosimeter response between batches was also studied.

1. INTRODUCTION

Radiation application in the field of environment, such as water purification and sewage treatment or vulcalnization of natural rubber, is a growing industry and an active developmental technology [1]. In Thailand, the study on the effect of gamma radiation on microorganisms in sludge from sewage treatment plant and hospital was carried out using gamma radiation in the dose range of 2 to 5 kGy [2] and the research on the improvement of natural latex by radiation have also been studied[3]. The sewage sample in a amount of millilitre was first treated in the laboratory and then transferred to a few hundred litres for industrial application. For process validation, the absorbed dose was measured by placing a commercially available dosimeters such as PMMA or nylon thin film dosimeters inside the sample container.

Oxalic acid dosimeter was first suggested by Draganic[4]. For oxalic acid (25-600 $mM \bullet L^{-1}$) dosimetry, the radiolytic mechanism is given as follows. With oxygen present, radiation decomposes an acid molecule producing two molecules of carbon dioxide(CO_2).

$$(COOH)_2 + OH \rightarrow CO_2 + H_2O + COOH$$
$$O_2 + H \rightarrow HO_2$$
$$COOH + HO_2 \rightarrow H_2O_2 + CO_2$$
$$HO_2 + HO_2 \rightarrow H_2O_2 + O_2$$

The CO_2 is a main product of the radiolysis of oxalic acid solution. Therefore the amount of decomposed acid is determined as the concentration difference between irradiated and unirradiated samples. Draganic introduced two methods which were titration with NaOH and spectrophotometric method for determination of absorbed dose which depended on the difference in concentration of oxalic acid. He also reported the radiation yield of oxalic(G_{ox}) which was 4.9 ± 0.4. The dosimeter is quite insensitive to impurities and very stable to normal storage before and after irradiation[5]. Holm[6] reported the process for determination of absorbed dose using oxalic acid by titration with NaOH. He proposed the initial oxalic acid concentration for different dose regions. The decomposition of dosimeter dose not proceed linearly with the absorbed dose.

The aim of the present work is to apply 2.5 mM oxalic acid as a liquid chemical dosimeter for absorbed dose measurement using a spectrophotometric method in various scale of sample solution. It is also to study the effect of various parameters following the criteria for selecting suitable dosimeter[7]. The studies for optimum wavelength selection, response characteristics, pre- and post-irradiation stability, reproducibility and the variation of dosimeter response between batches were carried out. The 100 mL and 8 L of oxalic acid solution was irradiation against Fricke in the same size of sample container for validation of the process.

2. EXPERIMENTAL

The oxalic dosimeter solution was prepared using analytical grade oxalic acid dihydrate ($H_2C_2O_4.2H_2O$; MERCK grade; M = 126.07 g•mole^{-1}) and diluted to 2.5 mM with distilled water. The dosimeter solution in 5-ml glass ampoules was irradiated in the centre position of a "Gamma-Cell 220" ^{60}Co gamma irradiator with annular geometry. The oxalic acid solution was calibrated against Fricke as a reference dosimeter. The absorbed dose rate at the time of irradiation was 3.12 Gy/s. The absorption spectra and optical absorbance at specific wavelengths were measured with a double-beam Shimadzu model UV-3101 PC spectrophotometer, using a band pass setting of 1 nm. The solution was held in the object beam in quartz-glass, 10 mm path-length cuvette, with the reference beam cuvette containing purified water. The 100 mL and 8 L of oxalic acid solution were filled in cylindrical flask (dimension: ∅ 0.5 x 7.5 cm and ∅ 0.23 x 36 cm) and were irradiated against Fricke with the same scale of solution containers using Gammabeam 650 ^{60}Co gamma irradiator .

3. RESULTS

The aqueous oxalic solution was irradiated for absorbed dose of 2.8, 5.6, 8.4 and 11.2 kGy. Fig.1 shows the optical absorption spectra of unirradiated and irradiated 2.5 mM of dosimeter solution over the range of 200-300 nm. The inset shows the decrease in the absorbance, A as a function of the absorbed dose. The optimum wavelength of 220 nm was selected. The optical density of the irradiated solution decreases with absorbed dose. The dose response in terms of net absorbance, $A_{unirrated}$-$A_{irirrated}$, is plotted as a function of absorbed dose.

In a study of the stability of the aqueous oxalic solution before and after irradiation for 2 kGy at the wavelength of 220 nm is shown in Fig.2. The variation in the optical density for storage over 25 days was less than 1% for unirradiated and ± 2% for irradiated solution. The dosimeter solution in glass ampoules were irradiated at the centre of the irradiation chamber 10 times at absorbed dose of 2 kGy for reproducibility study. The standard deviation of mean response was ± 1% (1 σ).

Figure 3 shows the variation of dosimeter response between batch A and B for the absorbed dose range of 1-6 kGy.

The 100 mL of oxalic acid solution was irradiated for 3.05 kGy against Fricke at dose rate 2.66 Gy/s in the same irradiation condition. The variation in the absorbed dose was within 3 %. The variation in the absorbed dose for 8 litre of oxalic solution and Fricke was ± 5% at 1.02 kGy and 1.18 Gy/s.

4. CONCLUSION

Oxalic acid (2.5 mM), whose density is about that of water, can estimate the absorbed dose in various size of containers over the dose range of 1-10 kGy by spectrophotometric analysis at 220 nm. The dosimeter solution should be calibrated against Fricke as reference dosimeter. The dosimeter solution can be prepared using normal distilled water.

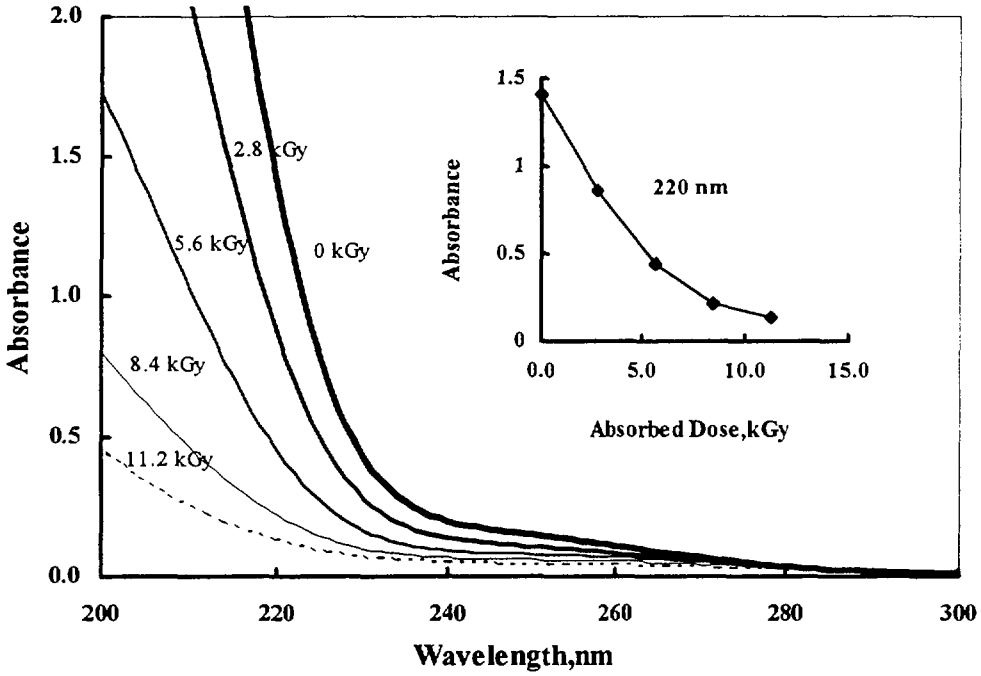

FIG. 1 Absorption spectra of unirradiated and irradiated aqueous oxalic acid; the insets show the decreases in absorbance at 220 nm

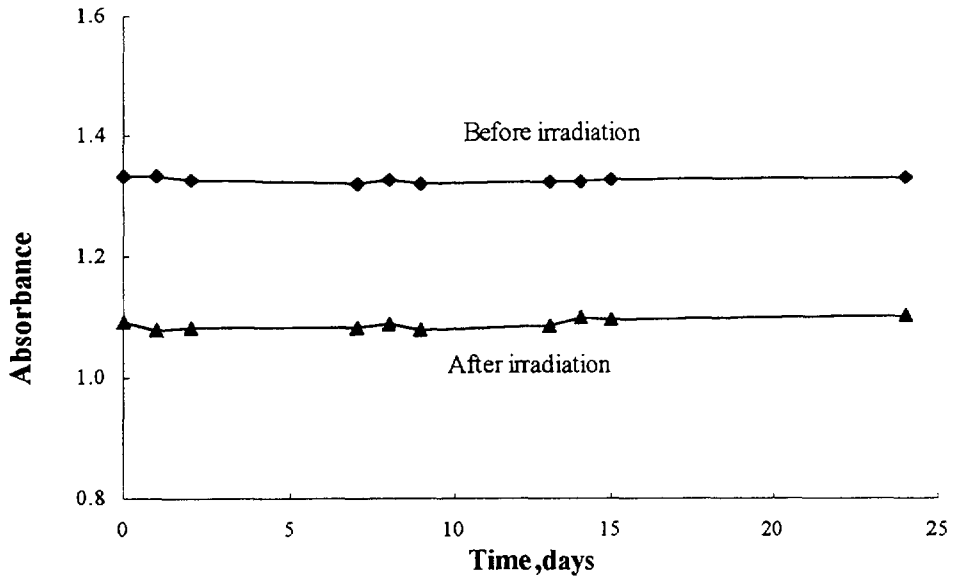

FIG.2. Stability in absorbance (λ= 220 nm) of unirradiation and irradiation 2.5 mM aqueous oxalic acid.

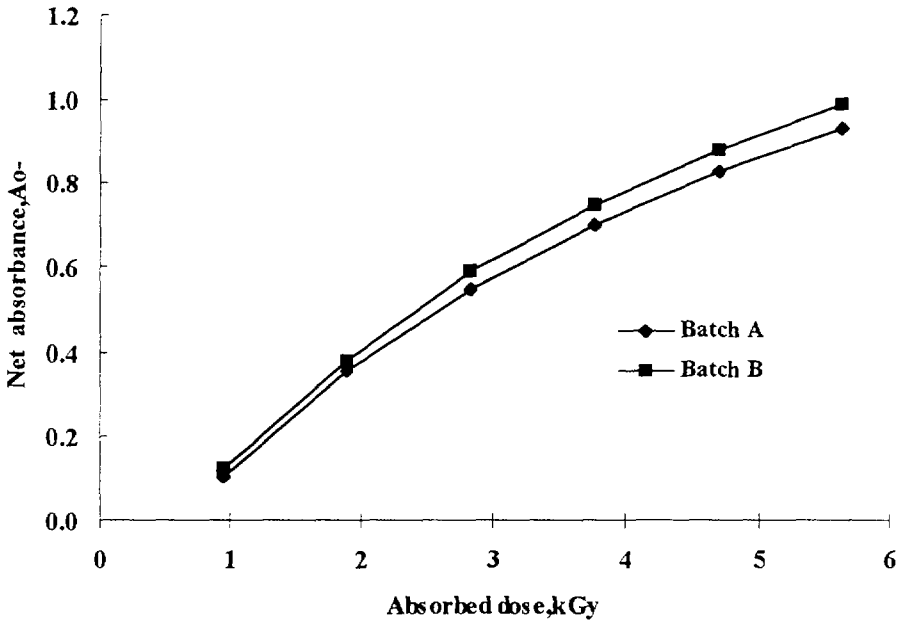

FIG. 3. Dose response curve of 2.5 mM aqueous oxalic acid batch A and batch B,measured at weavelength 220 nm

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