Dimethyl sulfoxide, diethyl fumarate solution for high dose dosimetry

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► ***Short report***

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ABSTRACT

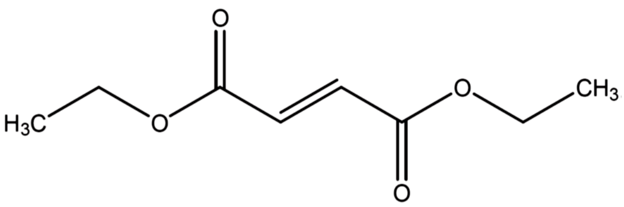
Background: Dosimetry is a fundamental part of the radiation processing industry. Aqueous inorganic dosimeters have been used for gamma radiation dosimetry such as standard ferrous sulfate dosimeter. The purpose of this study was to investigate the effect of gamma radiation on the diethyl fumarate DEF in dimethyl sulfoxide DMSO solution and its possible use in chemical dosimetry. ***Materials and Methods***: Three solutions having different concentrations of DEF were prepared (5, 10 and 20%) and exposed to various gamma doses. Irradiation was carried out using ROBO facility of ^{60}Co gamma ray. Absorbance was measured using Melton Roy 1201 UV-VIS spectrophotometer. ***Results:*** The absorption spectra of irradiated solution showed broad absorption bands between (325-400nm) with a shoulder at 332 nm. The absorbance increased with increasing the dose. Good response and linear relationship were found between absorbed dose and absorbance of 10% and 20% DEF concentration in the range of 0-225 kGy at the measured wavelength. Good dose rate independence was observed in the range of 14-33 kGy/h. It was noticed that irradiated samples had a good stability in darkness. Whereas, the response was affected during the storage in day light. This response was found to be depended on the irradiation temperature within the range of 18- 52 ^{0}C. ***Conclusion:*** Our measurements showed that the ability of using the solution of diethyl fumarate DEF in dimethyl sulfoxide DMSO as a high dose radiation dosimetry. The useful dose range for 10% and 20% DEF of concentrations is up to 225 kGy. Nevertheless, the correction of temperature for routine dosimetry measurements should be taken into account.

Keywords: *Diethyl fumarate (DEF), dimethyl sulfoxide (DMSO), gamma radiation, dose rate, post irradiation, dosimetry.*

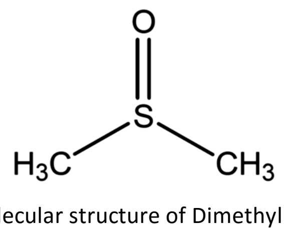
INTRODUCTION

Dosimetry is a fundamental part of the radiation processing industry [(1)]. Aqueous in organic dosimeters have been used for gamma radiation dosimetry such as standard ferrous sulfate dosimeter [(2)], ceric sulfate Ceric-Cerous Sulfate [(3)], Dichromate dosimeter [(4)], and Potassium Nitrate [(5)]. These dosimeters are well-established systems and have radiation-chemical mechanisms that are thoroughly understood. In practice, organic impurities in these dosimeters are still the main problem. Therefore, using organic chemical dosimeters eliminate the impurities problems such as methanol monochlorobenzene [(6,7)]. Also, most dosimeters based on organic compounds as solutions or polymer dyed films have wide range of doses response [(8-10)]. Radiation radiolysis studies on diethyl fumarate were carried out by few workers. It was reported that diethyl fumarate is dimerized during irradiation and absorbance at 335 nm was detected using pulse radiolysis technique [(11)]. In addition, the dosimetric characterization of diethyl fumarate DEF in dimethyl formamide DMF solution was studied by following the change of absorbance at 350, 360 and 370 nm after gamma irradiation in the range 1-125 kGy [12]. The results indicated that the concentration of 20% DEF had good linearity at 370 nm up to 125 kGy. Dimethyl sulfoxide (DMSO) was used as free radical scavenger in FPGX gel dosimeter system [13], and pararosniline cyanide in dimethyle sulfoxide solution was used also as chemical radiochromic dosimeter[14]. In the present work, a solution of diethyl fumarate DEF in dimethyl sulfoxide DMSO has been studied as a high dose radiation dosimetry in the range (0-225 kGy). Effects of dose rate, concentration of DEF, irradiation temperature, and post irradiation stability were also investigated.

Both diethyl fumarate DEF and dimethyl sulfoxide DMSO of 98% purity were purcha sed from FLUKA Company (Germany), and used without any additional purification (schemes 1 and 2). DEF solutions were prepared by dissolving 2.5, 5, 10 mL of DEF, respectively in volumetric flasks of 50 mL capacity, then diluted with DMSO to obtain solutions having concentration of 5%, 10%, 20% by volume.



Schemes 1. Molecular structure of Diethyl fumarate.



Schemes 2. Molecular structure of Dimethyl Sulfoxide.

MATERIALS AND METHODS

Materials

Both diethyl fumarate DEF and dimethyl sulfoxide DMSO of 98% purity were purchased from FLUKA Company (Germany), and used without any additional purification (schemes 1 and 2). DEF solutions were prepared by dissolving 2.5, 5, 10 mL of DEF, respectively in volumetric flasks of 50 mL capacity, then diluted with DMSO to obtain solutions having concentration of 5%, 10%, 20% by volume.

Irradiation of samples

Irradiation was carried out using the gamma facility Russian design type ROBO of ^{60}Co gamma ray with sources arranged in a rectangular holder with a total activity 185 kCi. All irradiation positions were calibrated using Fricke dosimeter. The samples for irradiation were prepared by taking 2.5 mL from each solution in glass ampoules, and then thermally were sealed. Irradiation of samples was carried out in polyethylene's boxes, of 3 mm thickness in air atmosphere at room temperature.

Instruments

Absorbance was measured using UV-VIS spectrophotometer type Milton Roy spectronic 1201 (supplied by the Milton Roy Company, USA). All spectrophotometric measurements were carried out at ambient temperature using glass cells with 1cm optical path length. Absorbance of the irradiation solution was calculated using the following relationship:

$$\textbf{Absorbance} = A - A_0$$

where A, A_0 are the absorbance of irradiated and non-irradiated solutions, respectively.

RESULTS AND DISCUSSION

The absorption spectra of solutions having concentration of 20% DEF were recorded using the non-irradiated solution as blank for all samples, which were exposed to doses up to 250 kGy at the dose rate of 30 kGy/h. The recorded spectra were shown in figure 1. The absorption spectra of irradiated samples showed absorption band in the range 320-400 nm and its maximal absorption was observed at a wavelength of 332 nm. It can be seen that the absorbance increased with increasing the irradiation dose. Any changes in the chemical structure of this molecule could cause a change of the solution absorbance. It was reported that diethyl fumarate is dimerized during irradiation and absorbance at 335 nm was detected using a pulse radiolysis technique [8]. Therefore, this increase in absorbance could be due to the formation of dimers.

Effect of concentration of DEF on the response of absorbed dose

Three solutions having different concentration of DEF: 5, 10 and 20%, respectively were prepared and then irradiated at dose rate of 30 kGy/h up to 250 kGy. Absorbance measurements were carried out 12 h after irradiation for all samples at wavelength 332 nm. The absorbance values at 332 versus the irradiation dose are reported in figure 2 for the three concentrations. Figure 2 demonstrates a good linearity relationship of absorbance at 332 mn in the rage 0-50 for the concentration 5%, while the linearity of response curves were extended up to 225 kGy for 10% and 20% DEF of concentrations. In a previous work, a dosimertric characterization of diethyl fumarate in dimethyl formamide was studied by Zayzafoon and Alkassiri 2008 who found a good linear absorbance at wavelength 370 nm up to 125 kGy. As seen from our results, the replacement of dimethyl formamide solvent by dimethyl sulfoxide extended the linear response up to 225 kGy.

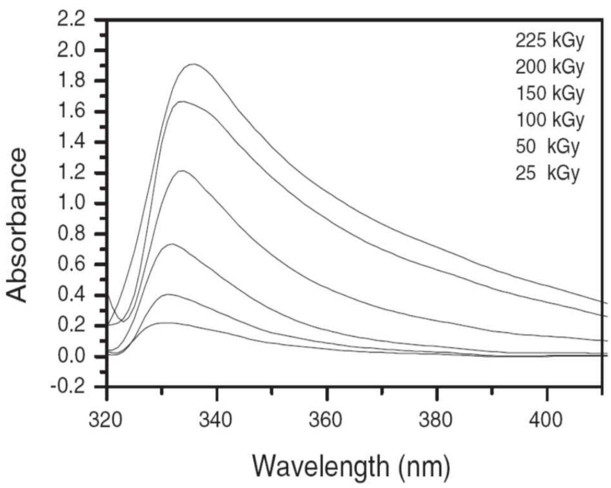


Figure 1. Absorption spectra of dimethyl sulfoxide diethyl fumarate for different irradiation doses (25-225 kGy) at concentration of 20% DEF and dose rate 30 kGy/h.

Figure 3 presents a linear regression of the response values of 20% concentration of DEF, which is represented by the following relationship:

$$A = 0.00838 \times D$$

Where A is the absorbance at 332 nm and D is the absorbed dose. These results suggest that the most useful concentration of DEF in DMSO is 20%. This finding is in agreement with a pervious work by Zayzafoon [9].

Post irradiation stability

An important influence quantity in ionizing radiation dosimetry is the time interval between

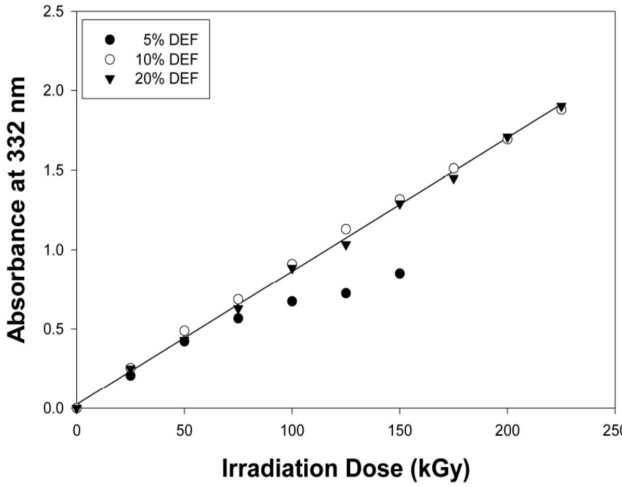


Figure 2. The absorbance values at 332 versus the irradiation dose for the concentration 5, 10 and 20% of DEF and at dose rate 30 kGy/h.

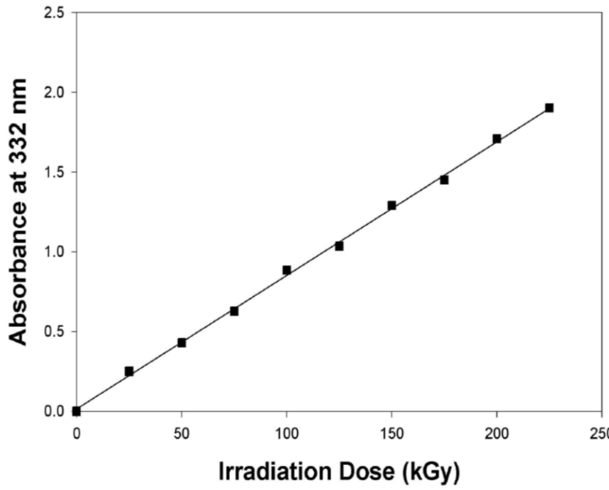


Figure 3. linear regression of the response values at 332 versus the irradiation dose for the concentration 20% of DEF and at dose rate 30 kGy/h.

irradiation and measurement of a dosimeter. This is a key consideration for dye-based radiochromic dosimeters that experience post-irradiation optical density changes with time [15,16]. So, the effect of storage conditions on the post irradiation stability of our dosimeter was investigated for solutions having concentration of 20% DEF. Two batches of solution were irradiated with 25 kGy and kept at room temperature at two conditions, one in darkness and the other in day light. Measurements at 332 nm were carried out periodically during the period of storage. Figure 4 shows the change of absorbance during storage. It was noticed that the irradiated samples had a good stability in darkness, and the change of absorbance during the storage had stability within 2% of absorbance change for over 30 days period of storage. Whereas, the change of absorbance after irradiation during the storage in day light was very clear during five days of storage and the change was more than 23% for 30 days of period storage.

Effect of dose rate

Most of established dosimeters were independence of dose rate, so the main step in selecting a suitable dosimetry system is the comparison of the various characteristics of the dosimetry systems with respect to irradiation conditions (such as temperature, dose rate and humidity) [17]. The effect of dose rate on the response of 20% concentration of DEF solution was studied at three dose rates (14, 24 and 30 kGy/h) in the dose range 0-225 kGy. Absorbance at 332 nm was measured and presented as shown in figure 5 .It can be observed that the dose response of three dose rates had the same behavior up to 225 kGy and exhibited good independency of dose rate, namely less than ±1% change in absorbance at the chosen dose rates, and they had the same linear relationship A = 0.00838 x D. This means the response of 20% DEF solution is independence of dose rate within the studied range of dose rate.

Effect of irradiation temperature

It is well known that the temperature plays an important role in the radiation processing [17]. Therefore, the effect of irradiation temperature on the response of dosimeter was investigated. Solutions having concentration of 20% DEF were irradiated to a dose of 30 kGy, dose rate 30kGy, at different temperatures (from 18 to 52°C) using thermostatic chamber within ±2 °C precision. The absorbance at 332 nm versus the temperature was measured and given in figure 6. It can be seen that the absorbance of 20% DEF increase linearly with the temperature. The temperature correction was calculated against the absorbance of solution irradiated at 25 °C

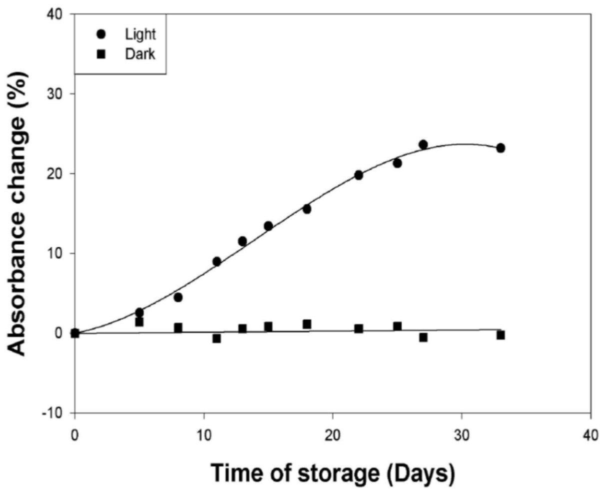


Figure 4. Post-irradiation stability of dimethyl sulfoxide diethyl fumarate irradiated for the concentration 20% DEF in dark and indirect daylight at ambient temperature.

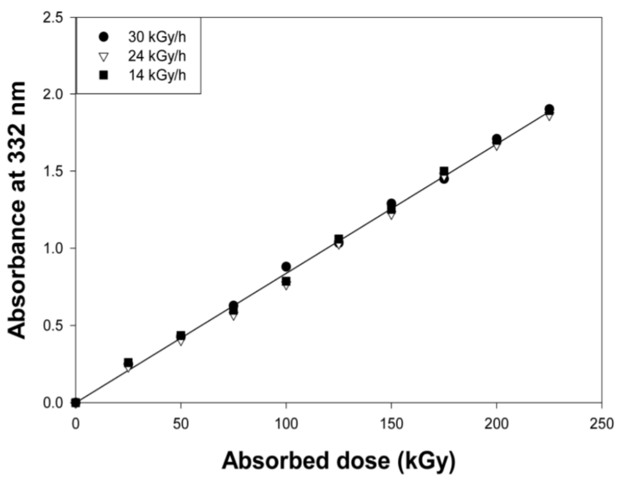


Figure 5. The absorbance values at 332 versus the irradiation dose for the concentration 20% of DEF and at dose rate 14, 24 and 30 kGy/h.

and given by the following empirical equation:

$$A_{25} = \frac{A_t}{1 + 0.0172(t - 25)}$$

Where A_{25}: Absorbance of solution irradiated at 25 °C.
A_t: Absorbance irradiated at temperature t
t: Irradiation temperature.

The correction of temperature for routine dosimetry measurements should be taken into account. It can normalize the absorbance of solution irradiated at (t) temperature to the absorbance irradiated at 25 °C.

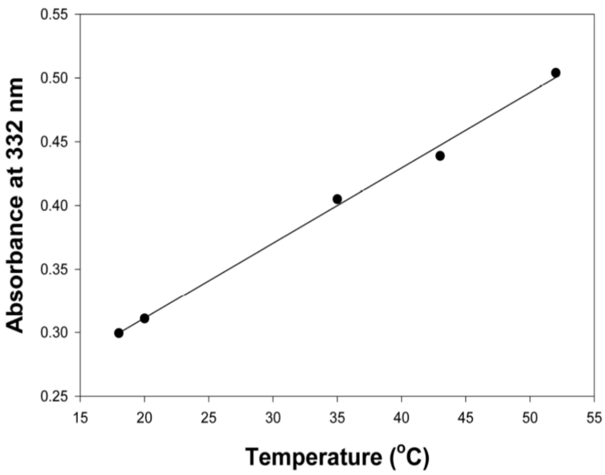


Figure 6. The absorbance values at 332 versus the irradiation temperature for the concentration 20% of DEF irradiated at 30 kGy and at dose rate 30 kGy/h.

CONCLUSION

Various solution compositions of DEF and DMSO were investigated. The results showed a good linearity relationship of absorbance at 332 nm in the range 0-50 for the concentration 5%, while the linearity of response curves were extended up to 225 kGy for 10% and 20% DEF of concentrations, and exhibited a good independency of dose rate. The irradiated samples had a good stability in darkness, within 2% of absorbance change for over 30 days period of storage. Whereas, the change during the storage in day light was very clear during five days of storage and the change was more than 23% for storage period of 30 days of period storage. Nevertheless the absorbance of 20% DEF increased linearly with the irradiation temperature and correction of temperature for routine dosimetry measurements should be taken into account.

ACKNOWLEGMENTS

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Conflicts of interest: *none to declare.*

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