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Analysis by EPR measurements and spectral deconvolution of the dosimetric properties of lithium formate monohydrate

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ABSTRACT

In this study we analyze the Electron Paramagnetic Resonance “EPR” response of lithium formate monohydrate “LFM” irradiated by photons and electrons using a linear accelerator. EPR measurements were performed one day after irradiation and regularly during six months of storage. Both dosimetric curves using peak to peak technique and double integration method showed linear behavior. The time dependence indicates good stability of free radicals produced on this material. The determined measurable threshold dose is 0.55 Gy. For used energies, LFM dosimeters are more sensitive to X-ray irradiation than to electrons. The deconvolution of the EPR signal of irradiated LFM using Levenberg-Marquardt (M-L) algorithm shows that this spectrum is constituting of three gaussian contributions. Each contribution has a linear evolution according to the irradiation dose, as well as an acceptable degree of stability during the storage period, approving the stability of all free radicals produced in LFM. All these characteristics confirm that LFM might be a promising dosimeter to be used in the ‘clinical’ dose range in radiotherapy.

1. Introduction

Electron paramagnetic resonance is a helpful technique for dosimetric purposes, it allows obtaining information on the irradiated material by measuring the concentration of free radicals produced by ionizing radiations [1]. This advanced technique is precise and sensitive. The non-destructive readout and the possibility to perform cumulative dosimetry are the most attractive properties of this technique [2–5].

Main features of a material to be a suitable dosimeter are the linearity and the sensitivity of its response to irradiation dose, its lower limit of detection and its stability over a long-time period after irradiation [6]. Several materials have been investigated, among them, alanine [7–9], table sugar [10–12], formates and dithionates [13–17].

Lithium formate monohydrate (LFM) is another promising material consists of a single broad line and its radical formation under irradiation was well studied by EPR spectroscopy. This material respond linearly to absorbed dose [16,18,19] and exhibit low fading effect under optimal atmospheric conditions during storage period after irradiation [18,20]. Among the radicals constituting the spectrum of LFM, $^{\cdot}CO_2^{-}$ radical was observed as the dominant species of this substance [19,21,22].

The aim of this work is to enlarge the study by EPR of LFM spectrum irradiated by 6 and 18 MV X-rays photons and by 6 and 18 MeV electrons. For this purpose, in the first part, we analyzed the dosimetric properties of the entire EPR spectrum according to the delivered dose (0–20 Gy) in order to check the linearity of dosimetry curves and to determine the measurable threshold dose. The energy dependence of this material is also evaluated and the fading effect is inspected during a storage period of six months after irradiation.

The main part of this work is conducted on a deconvolution study of LFM spectrum in order to identify the contributions constituting this spectrum and to analyze the behavior of each contribution according to the attributed dose and to the storage time after irradiation. For this purpose, Levenberg-Marquardt algorithm is utilized on the integral absorption spectrum (obtained after integrating the experimental EPR spectrum while considering a baseline correction [11]) of LFM using different adjustment functions (Gaussian, Lorentzian and Voigtian) with varying their number of lines [23–25]. The choice of the number of contributions and their representative function is based on three criteria, a better fit between the integral absorption spectrum with the spectrum obtained by summing the different contributions, the different contributions must present a homogeneous evolution as a function of the irradiation dose and finally they must provide a homogeneous behavior as a function of the storage time after irradiation. Both peak to peak amplitude of the EPR measured spectrum and the

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Table 1
Spectrometer settings used for EPR measurements.

Parameter	Optimal value
Room temperature	23 ± 1 °C
Cavity temperature	30 ± 1 °C
Microwave power	1 mW
Modulation amplitude	0.5 mT
Sweep time	60 s
Spectrum resolution	4096 points
Time constant	0.1 ms
Number of scans	1–20 times

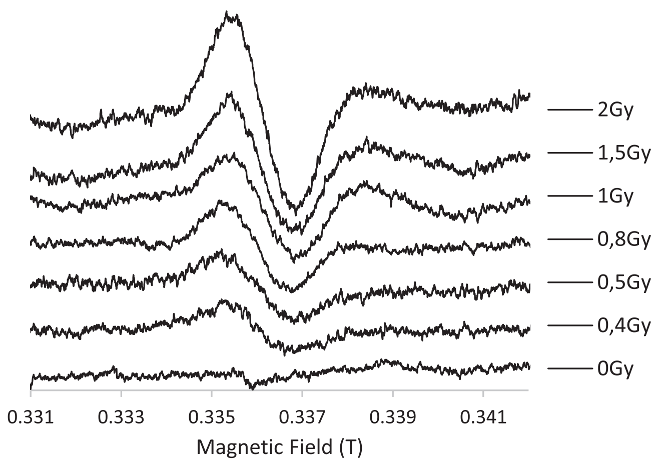

Fig. 1. First-derivative EPR spectra of LFM dosimeters irradiated by X-6 MV to different doses [0.4–2 Gy] using the optimal parameters.

value of area under the integral absorption signal were used in the different parts of this study.

2. Instruments and methods

2.1. Irradiation and measurement

Lithium formate monohydrate powder was purchased from Sigma-Aldrich, United States. This material was irradiated by 6 and 18 MV X-rays photon beams (X-6 and X-18) and also by 6 and 18 MeV electron beams (E-6 and E-18) using a linear accelerator "VARIAN – Clinac DHX" installed at Al Kindy oncology center in Casablanca, Morocco. Samples of 500 mg were filled inside capsules (Radius = 14 mm; Height = 4.8 mm) to be irradiated in the dose interval 0–20 Gy. Irradiations were performed placing these samples inside a PMMA phantom at depths: 1.4 cm for X-6, 3.3 cm for X-18, 1.2 cm for E-6 and 3 cm for E-18. These depths correspond to the maximum dose of each used particle and energy. They were established by performing dose measurements at different depths in a water phantom using an ionization chamber. The field X-Y of the irradiation is 10 × 10 cm^2 and the distance from irradiation source to phantom surface is fixed at 100 cm. Two dosimeters were irradiated for each energy and for each dose at the same moment.

For the investigation of time dependence. Several dosimeters were irradiated by X-6 (doses: 2, 3, 6, 10, 15 and 20 Gy) and kept in well-sealed tubes under ambient conditions, to be regularly analyzed over a period of six months.

EPR measurements were undertaken on irradiated samples of LFM (weight: 200 ± 0.5 mg) using a Magnettech MS400 (Berlin, Germany) spectrometer operating at the X-band. The measurement parameters are presented in Table 1. Microwave power was chosen in the linear response region in order to avoid the saturation of signal and optimize the signal-to-noise ratio. A reference sample of 1,1-diphenyl-2-picrylhydrazyl (DPPH) was used as a standard reference for the calibration of the spectrometer.

2.2. Treatment methods

Peak-to-peak amplitude and double integration method have been exploited in this study to analyze the EPR spectrum of irradiated LFM, according to the irradiation dose, storage time, and energy effect. A deconvolution computation, based on Levenberg-Marquardt algorithm, is conducted on the integral absorption spectrum in order to determine the most probable nature of the lines constituting this spectrum. Different attempts are evaluated by testing many combinations of different functions (Gaussian, Lorentzian and Voigtian). In this study, deconvolution calculations were carried out using OriginLab program.

3. Results and discussion

3.1. EPR measurement results

3.1.1. EPR spectra and dose response

For each used dose, two samples were irradiated and four EPR measurements were undertaken. Fig. 1 shows the EPR spectrum of

Table 2
The relative standard uncertainties calculated for the curves of LFM irradiated by X-6 MV and obtained 24 h after irradiation.

Dose (Gy)	Peak-to-peak amplitude			Double integral		
	Mean (a.u.)	Std. deviation	Estimated error (%)	Mean (a. u.)	Std. deviation	Estimated error (%)
0	197	25	12	–	–	–
0.4	326	17	7.11	2.8E−04	3.12E−05	7.82
0,5	425	18	4.86	3.1E−04	1.85E−05	5.72
0.6	472	17	3.78	394E−04	1.53E−05	3.43
0.8	517	20	3.49	4.5E−04	1.62E−05	3.17
1	635	27	4.83	5.59E−04	3.44E−05	5.02
1,5	808	41	4.92	7.87E−04	5.12E−05	3
2	1219	81	4.12	10.74E−04	5.7E−05	4.48
2,5	1475	51	3.36	12.25E−04	3.49E−05	2.76
3	1837	25	1.34	15.64E−04	7.10E−06	0.44
3,5	2226	104	4.63	18.59E−04	8.73E−05	3.41
6	4064	165	4.04	33.09E−04	15.33E−05	1.07
10	6705	112	1.65	56.12E−04	15.34E−05	2.22
15	9671	193	1.98	80.75E−04	18.85E−05	1.19
20	12.875	342	2.63	107.6E−04	30.13E−05	2.71