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EPR dosimetric properties of radiation – Formed radicals in arginine monohydrochloride

W.B. Beshir *, A.A. Abdel-Fattah, F. Abdel-Rehim, H.M. Hassan

National Center for Radiation Research and Technology, P.O. Box 29, Nasr City, Cairo, Egypt

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ABSTRACT

Arginine monohydrochloride rods (3 × 10 mm) were irradiated with ^{60}Co γ-rays to study radicals for dosimetric materials with Electron Paramagnetic Resonance (EPR). The rods have significant signal which develops upon irradiation and the intensity of signal increases upon the increase in irradiation dose. The rods can be used in the dose range from 5 to 120 KGy. The temperature coefficient was found to equal +0.22% °C^{-1}. The dose response, influence of humidity and post-irradiation storage at different conditions are discussed. The overall uncertainty for calibration of arginine monohydrochloride rode dosimeters at 2σ was found to be 2.85%.

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1. Introduction

A number of solid materials, including amino acids, in which free-radical populations are formed by irradiation, have been suggested for high-dose dosimetry by Electron Paramagnetic Resonance (EPR) analysis. A useful method of high dose measurement is the use of EPR spectrometry of irradiated amino acids, in particular L-alanine, $CH_3CH(NH_2)COOH$, purified in polycrystalline form [1–4].

Regulla et al. [5] have developed a dosimeter that can be used with 3.5% (1σ) precision limits, even when mailed over large distances and used under extreme climates, with several months between irradiation and analysis. The dosimeters have been made as small rods 4.9 mm diameter and 10 mm length, consisting of ~90% L-alanine by weight suspended in ~10% paraffin. A system with different formulations and packaging has been described [6]. The amino acid has also been suspended in other hydrocarbon binders, such as cellulose or polyvinyl pyrrolidone [7–9] or polystyrene [10,11] or it can be protected by a painted layer [12] or produced as a thin polymer film containing alanine [10,11,13].

In 1989 pellet-shape alanine dosimeter (0.95 mass fraction of L–α–alanine and 0.5 mass fraction of polyvinyl-pyrrolidinone) have been prepared [14]. In 1991 at the National Physics Laboratory (NPL) alanine dosimeter pellets [15] 5 mm diameter and 2.5 mm thickness with average weight 55 mg using 90% alanine and 10% paraffin wax by weight were prepared. A pellet shape dosimeter was also prepared (0.7 mass fraction of DL–α–alanine and 0.3 mass fraction of low density P.E.) [16,17].

A polymer alanine film to be used for dosimetry of ionizing radiation was introduced in 1993 [18]. The film thickness was 0.3–0.4 mm. The films show good linearity of dose response in the absorbed dose range 0.1–10 kGy.

A new alanine dosimeter in 2005 was prepared [19] using poly (vinyl butyral – co – vinyl alcohol – co – vinyl acetate) copolymer as a binder with ratio 40% L-alanine to 60% binder by weight in the form of pellets 5 mm in diameter and of average height 3.5 mm.

Several improvements to reduce the orientation effects can be accomplished by certain time-consuming procedures, e.g. for pellet-shaped dosimeters, averaging the measured values at two different orientations 90° apart [20,15] or by using double-integration of the spectrum [21], and for film dosimeters, by attaining maximum amplitude or by averaging the maximum and minimum amplitudes [22].

The objective of the present study is preparation of new EPR dosimeter by a simple technique in the laboratory through mixing arginine monohydrochloride with a mixture of vinyl acetate copolymer and paraffin wax. The dose response, effects of humidity and temperature as well as pre- and post-irradiation stability are discussed.

2. Experimental

2.1. Materials, instruments and methods

2.1.1. EPR spectrometer and parameters

The system used in this study is an EMX-BRUKER-system-Germany, supplied by a 9.5 GHz microwave (X-band) Gunnoscillator Bridge with automatic tuning capability and a rectangular 4102 ST cavity operating in the TE102 mode. The standard EMX Signal

* Corresponding author.
E-mail address: Dr.wafaa@live.com (W.B. Beshir).

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Channel can be operated at any modulation frequency between 6 kHz and 100 kHz, and has unsurpassed phase resolution and stability.

2.1.2. Radiation source and radiation dose determination

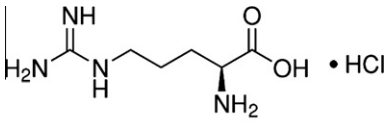
Irradiation was performed using a ^{60}Co gamma source (irradiation cell) installed at the National Center for Radiation Research and Technology (NCRRT) of the Egyptian Atomic Energy Authority. The source activity was 50,000 Ci, with a dose rate of about 6.078 kGy/h at sample position. Samples were irradiated at room temperature and the radiation doses delivered to samples range from 5 kGy to 120 kGy.

2.2. Evaluation method

Empty tube spectra were measured before recording sample spectra in order to ensure the purity of the obtained signals. Readings were corrected to the peak-to-peak height of the reference standard material (DPPH, α, α-diphenyl β-picrylhydrazyl), which was recorded before and after each signal spectrum of the samples in order to correct for the change in the spectrometer sensitivity.

Sample mass normalization was performed for each acquired signal intensity. Each spectrum is of a single scan (n = 1).

L-arginine monohydrochloride, (11040, BioChemika, Assay ⩾99.0% (AT)) decomposition temperature >200 °C, hot melt stick adhesive based on ethylene vinyl acetate copolymer (EVA) (TEC-Bond 232/12, Power Adhesives Limited, England) and Paraffin Wax (Congealing point 65–71 °C, BDH), have been used for the rods preparation.



Arginine Monohydrochloride

2.3. Preparation of arginine monohydrochloride rods

An equal weight mixture of paraffin wax and EVA was melted at 85–95 °C in a water bath. 5%, 10%, 15%, 20%, 25%, 30%, 40% and 50% fine powdered arginine monohydrochloride material was added to the hot mixture solution and mechanically stirred for about 15 min at the same temperature to obtain a homogeneous mixture. The hot solution is sucked into polypropylene tubes (inner diameter 3 mm) and was left to solidify by cooling. Arginine monohydrochloride mixture rod was obtained by removing the polypropylene tube then cut into rods (3 × 10 mm dimensions). The average mass of the prepared rods was found to be 0.07 ± 0.005 g.

2.4. Irradiation of the prepared rods

The Gamma cell 220 Excel ^{60}Co irradiation facility was used for irradiation of the prepared rods. The absorbed dose rate was about 6.078 kGyh^{-1} overall the time of the experimental part. Eight different types of rods were prepared depending on arginine monohydrochloride concentration. Five rods at each concentration were irradiated together at the central position of the sample chamber using a specially designed holder made from polystyrene to ensure electronic equilibrium.

EPR signals were recorded at room temperature by using a Bruker EMX spectrometer (X-band) product of Bruker, Germany. The operating conditions are, microwave power = 2.533 mW, modulation amplitude = 3.00 Gauss, modulation frequency = 100 kHz, sweep width = 300 Gauss, microwave frequency = 9.779 GHz, time constant = 163.84 ms and conversion time = 40.96 ms. The bottom of the EPR tube was adjusted at a fixed position to ensure reproducible and accurate positioning of the rods in the sensing zone of the cavity.

EPR spectra were recorded at two orientations of each rods in the EPR cavity (0° and 90°). The dose responses of dosimeters were calculated in terms of average peak-to-peak heights of the two orientations (h_0 and h_{90}) per unit weigh of dosimeter and normalized to the receiver gain of the EPR spectrometer. Stability of EPR spectrometer sensitivity was checked before and after each series of measurement using reference irradiated alanine dosimeters.

3. Results and discussion

3.1. EPR spectra recorded for arginine monohydrochloride rods of different concentrations

Different concentrations (5%, 10%, 15%, 20%, 25%, 30%, 40% and 50%) of arginine monohydrochloride rods were irradiated to a dose of 25 kGy. An EPR signal begins to develop upon irradiation and its intensity increases with the increase in concentration while the unirradiated arginine monohydrochloride rod showed no EPR signal (Fig. 1).

EPR spectrum of irradiated arginine monohydrochloride sample, shows more than one peak which suggests the presence of more than one paramagnetic center present in the sample as shown in Fig. 2. Signal amplitude with g = 2.018 (Peak 1) was chosen to perform all the work in this study (g is the g factor which is proportionately constant approximately equal to 2 for most sample, but varies depending on the electronic configuration of radical or ions in the sample).

The EPR spectra of arginine monohydrochloride rods (concentration 15%) was recorded after irradiation of rods to doses 5, 7, 10, 15, 20 and 25 kGy as shown in Fig. 3. It can be seen that the EPR signal begins to develop upon irradiation and its amplitude increases gradually with increasing the absorbed dose of γ-ray photons without any change in its shape.

3.2. Dose response

Eight sets of arginine monohydrochloride rods of concentrations (5%, 10%, 15%, 20%, 25%, 30%, 40% and 50%) were prepared and irradiated to different doses of γ-radiation in the range from 5 to 180 kGy. ESR spectra were recorded to establish the response curves (each dose point is represented by the average value of 5 rods measurement).

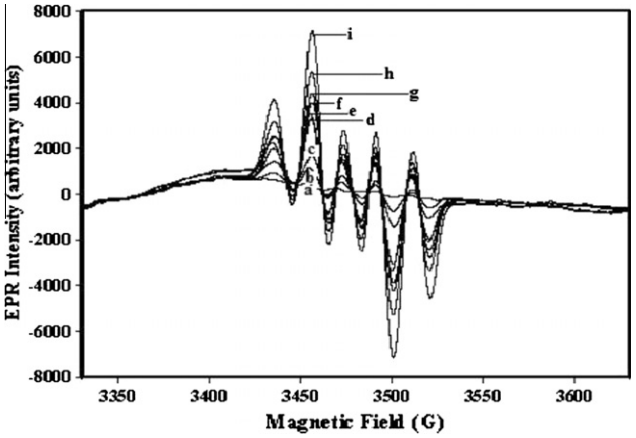


Fig. 1. EPR spectra recorded for arginine monohydrochloride rods of different concentrations irradiated to a dose of 25 kGy, (a) non-irradiated, (b) conc. = 5%, (c) 10%, (d) 15%, (e) 20%, (f) 25%, (g) 30%, (h) 40%, and (i) 50%.

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