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Resolving the limitations of using glycine as EPR dosimeter in the intermediate level of gamma dose

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ABSTRACT

The dosimetric properties of the simplest amino acid "glycine"- using EPR technique- were investigated in comparison to reference standard alanine dosimeter. The EPR spectrum of glycine at room temperature is complex, but immediately after irradiation, it appears as a triplet hyperfine structure probably due to the dominant contribution of the ($\bullet CH_2COO^-$) radical. The dosimetric peak of glycine is at g-factor 2.0026 ± 0.0015 and its line width is 9 G at large modulation amplitude (7 G). The optimum microwave was studied and was found to be as alanine 8 mW; the post-irradiation as well as the dose rate effects were discussed. Dosimetric peak intensity of glycine fades rapidly to be about one quarter of its original value during 20 days for dried samples and it stabilizes after that. The dose response study in an intermediate range (2–1000 Gy) reveals that the glycine SNR is about 2 times more than that of alanine pellets when measured immediately after irradiation and 4 times more than that of glycine itself after 22 days of irradiation. The effect of energy dependence was studied and interpreted theoretically by calculation of mass energy absorption coefficient. The calculated combined uncertainties for glycine and alanine are nearly the same and were found to be 2.42% and 2.33%, respectively. Glycine shows interesting dosimetric properties in the range of ionizing radiation doses investigated.

1. Introduction

Electron paramagnetic resonance (EPR) dosimetry is an important branch as an indirect ionizing radiation dosimetry. EPR dosimeters can be classified as organic like alanine (Bradshaw et al., 1962), inorganic materials like micro- and nano-barium sulfate (Aboelezz et al., 2015a), and metal ion-organic like magnesium lactate (Hassan and Ikeya, 2000). The advantage of the dosimetry using organic materials is closer to the radiation interactions that affect the cells in the living organisms rather than inorganic materials (Regulla and Deffner, 1982). The most prominent organic dosimeter is alanine, one of the non-polar amino acids, which is considered as reference of EPR dosimetry in intermediate and high dose ranges of radiation. Many articles were published on the dosimetric properties and applicability of alanine dosimeter especially in high dose range (Schneider et al., 1985; Mehta and Girzikowsky, 1996; ISO, 2013; Nagy et al., 2000; Malinen et al., 2003a, 2003b). Bartolotta et al. (1999) studied the EPR response of some different amino acids to ionizing radiation and reported that L-alanine has the highest response.

Both alanine and glycine are originated from the same group of amino acids, non-polar; alanine is nonessential amino acid while glycine is considered an essential one. The latter has the smallest chain of non-polar amino acids (NH_2CH_2COOH). Among this group, it has the simplest EPR spectrum with the hyperfine splitting ratio 1: 2: 1. Slifkin et al. (1984) compared to the dose response between alanine and glycine in ultra-dose range (0.1–2.5 MGy) from Co-60 source. Some researchers investigated the effect of different heavy ions irradiation on glycine using EPR (Henriksen, 1966; Weidong et al., 1998).

To the best of our knowledge, the radiation induced signal from glycine has not been studied till now from the view point of dosimetry. Thus, we will focus in this article on the dosimetric properties of glycine compared to alanine. The possibility and the feasibility of its use as EPR dosimeter in radiotherapy range of radiation dose will be also discussed.

2. Materials and methods

Polycrystalline glycine samples were purchased from S. D. Fine Chemical Limited India, and dried before irradiation at 120 °C for 0.5 h to remove their water contents. The irradiation process occurred at room temperature (22 ± 2 °C) to avoid the effect of irradiation temperature (Henriksen, 1966; Regulla and Deffner, 1982). The exposed samples were stored at dark area in tightly closed container. They are irradiated to 2 Gy from different beam qualities, radiation sources with

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diverse energies, that is, ^{60}Co, ^{137}Cs, and X-ray with different HVLs, to study the energy dependence of glycine. The X-ray tube model is MCN-323 metal-ceramic Philips double pole. The applied voltages were 70, 100, and 135 kV with constant tube current (10 mA). The doses are measured using NIS secondary standard system that is traceable to SI unit (calibrated at BIPM). For X-ray irradiation, glycine was exposed to X-ray after adding the additional filters that achieve HVLs which were recommended by the Consultative Committee of Ionizing Radiation (CCRI) of the Bureau International des Poids et Mesures (BIPM) (TRS 457 (Technical Report Series (TRS), 2007)). Moreover, the dose response of glycine is compared with reference EPR dosimeter, alanine pellets (Bruker BioSpin, Germany), through irradiating them to various doses of Co-60 gamma ray in the intermediate dose level from 2 Gy to 1 kGy. Three samples were irradiated for each dose; then they were stored in a dark area.

EPR spectra were measured, immediately after irradiation, by X-band EPR spectrometer (Bruker, EMX) at room temperature using high sensitive standard cylindrical resonator (ER 4119HS) operating at 9.7 GHz, with a 100 kHz modulation frequency. The maximum signal-to-noise ratio for signal was provided by choosing the optimum EPR parameters. These parameters are 8 mW microwave power for glycine and alanine pellets, the modulation amplitude is 7 G and 5 G for glycine and alanine, respectively, and the response time constant was 20 ms with a sweep time of 84 s. The weight of glycine was around 100 mg to fit 10 mm height inside the quartz tube.

3. Results and discussions

3.1. EPR spectrum

Fig. 1(a, b) demonstrates the radiation induced signal of glycine that was measured within a couple of hours after irradiation to 50 Gy from ^{60}Co source at room temperature. An isotropic hyperfine structure that is composed of three distinct peaks with theoretical relative intensities 1: 2: 1 is distinguished, while the experimental ratio of these intensities was 1: 2.5: 1.25. The separation between each two spectral lines is defined by the hyperfine coupling constant and was found to be $a = 20.3 \pm 0.4$ G (56.90 MHz). This structure is attributed to the interaction between nearby protons, with nuclear spin ($I=1/2$), and unpaired electrons in glycine radical ($\bullet CH_2COO^-$) producing triplet hyperfine pattern (Weiner and Koski, 1963; Morton, 1964). Recently, Talbi et al. (2004) have identified a triplet of triplet with $a_N = 15.1$ G doublet and $a_H = 3.3$ G and also a triplet of $a_N = 14.7$ G and $a_H = 7.9$ G. The signal at g-factor 2.0026 ± 0.0015 can be used as dosimetric peak, which is characterized by linear dose response. There is a slight

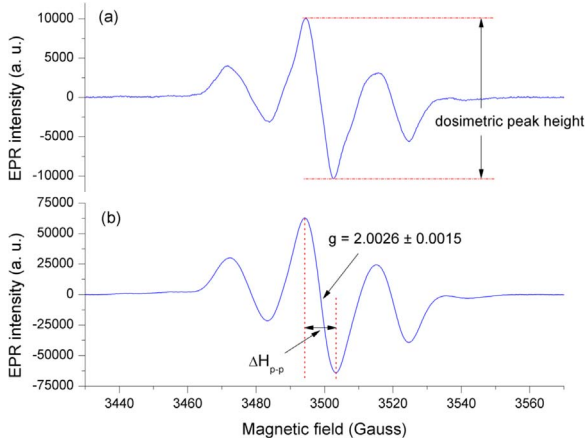

Fig. 1. EPR spectrum of glycine was measured directly after irradiation to 50 Gy from ^{60}Co source at g-factor 2.0026. The resulting $\Delta H_{p\text{-}p}$ of dosimetric peak is equal to 8 G for (a) at modulation amplitude 1 G and 9 G for (b) at modulation amplitude 7 G.

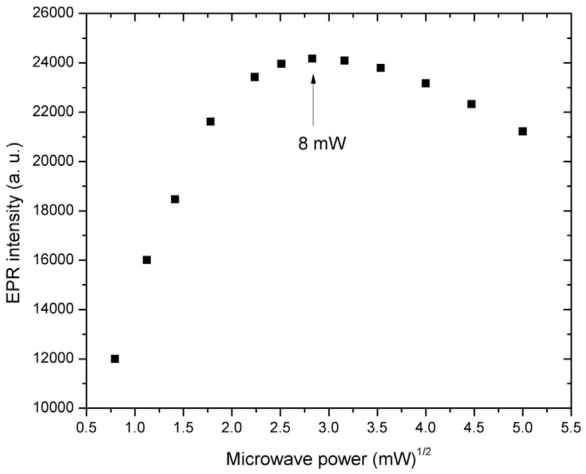

Fig. 2. The relation between EPR intensity and different microwave powers. The optimum microwave power is 8 mW.

difference in the peak-to-peak width ($\Delta H_{p\text{-}p}$) of dosimetric signal between well-resolved signal at 1 G modulation amplitude and over-modulated signal at 7 G in Fig. 1(a) and (b), respectively. It increases from 8 G to 13 G with the increasing of modulation amplitude from 1 (Fig. 1a) to 17 G, while the width was 9 G at modulation amplitude 7 G as shown in Fig. 1(b).

3.2. Microwave power dependence

A proper selection of microwave power is to reach the maximum ratios of "dosimetric signal-to-noise" and "dosimetric signal-to-native" (TECDOC-1331; IAEA, 2002). Fig. 2 portrays the dependence of EPR intensity on applied microwave power. It is clear from the figure that EPR intensity of irradiated glycine, which was measured directly after exposure, grows up rapidly with the increase of microwave power up to 8 mW and then it falls down slowly. Therefore, the optimum microwave power to be used is 8 mW.

3.3. Fading

The variation of EPR spectra of glycine between the direct measurement after irradiation to 50 Gy gamma rays and the measurement after 64 days, stored at room temperature, is shown in Fig. 3(a) and (b), respectively. It is observed that the height of the two side signals fades more rapidly than the height of the dosimetric peak. Sixty-four days after irradiation, the experimental intensities ratio of the triplet hyperfine structure changes to 1: 5: 1.25. Also, it is noticed that the line width of dosimetric peak increases as the post-irradiation time increases till 20 days. It belays after 20 days and it was found to be 12.5 G at large modulation amplitude (7 G).

Fig. 4(a and b) highlights the fading (post-irradiation) effect on EPR intensity of dosimetric signal for the dried and the undried glycine, respectively. It is obvious that they decrease exponentially and reach about one-fourth of their original values which is nearly consistent with the fading percentage of irradiated glycine by nitrogen ion that is reported by Weidong et al. (1998). The growing down stops and EPR intensities stabilize after 20 days for dried sample (Fig. 4a) and 30 days for the undried one (Fig. 4b). This fading attitude may be attributed to the intermolecular exchange that is predicted by Morton (1964). Larry et al. (1975) confirmed that a significant intermolecular exchange occurs within a few hours at room temperature and was clearly fast. In contrast to glycine, they hardly noticed an exchange in L-alanine after a month from irradiation. However, the exponential rate decay of ($\bullet CH_2COO^-$) radicals, the remaining from these radicals (~ 25% from the original concentration), is stable at room temperature which is in an