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Can Ammonium Tartrate Replace alanine in EPR Radiation Dosimetry?

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Abstract: EPR is one of the most recent and accurate techniques for radiation doses measurements, which is characterized by non-destructive evaluation of the radiation-induced radicals. Alanine is considered as the reference EPR dosimeter for several applications over decades due to its consistent response and the stability of its radiation-induced radicals. Recently, ammonium tartrate was proposed as an EPR promising dosimeter as it possesses several prominent dosimetric features.

In this work, ammonium tartrate is being investigated as a possible alternative to alanine. Studied properties included the sensitivity to different radiation doses, energy dependence, detection limit, and the stability of the induced radicals. Response to Cs-137 gamma radiation was studies and compared to alanine over two ranges, the first ranged from 47 to 2500 Gy, and the second ranged from 1.46 to 87.8 Gy. Uncertainties associated to the evaluated radiation doses using EPR/ ammonium tartrate dosimetry system were evaluated and presented in details.

Keywords: Radiation dosimetry; alanine; ESR; EPR; ammonium tartrate

1. Introduction

Electron paramagnetic resonance spectroscopy (EPR) evaluates the unpaired electrons in materials and can be employed for the measurements of radiation doses. Alanine was first proposed as radiation dosimeter in 1962 [1], and since that date it is considered as the reference EPR dosimeter for several applications of ionizing radiation, this may due to the exceptional dosimetric features of alanine: the high stability and the wide range of proportionality to radiation doses especially for high doses, and energy response which matches the human soft tissue properties in addition to its non-toxicity as it is an amino acid [2].

However, there are some drawbacks disabled the extension of alanine dosimetry to modern medical applications, these features include its complicated EPR spectrum which is attributed to three different radicals at least [3], also, its complex time dependence which varies with the level of applied radiation doses [4], in addition to the limit of detection which is hardly can reach values lower than 2 Gy [5]. Several methods were used in order to increase the sensitivity of alanine to lower doses, such as addition of nanoparticles [6], use of digital filters [7], and the use of very complicated impractical experimental procedures [8].

Several materials were proposed as possible EPR dosimeter [9-14], one of these

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proposed material is the ammonium tartrate which proved through extensive studies the promising spectroscopic and dosimetric features: simple EPR spectrum, highly stable radiation-induced radicals and lower limit of detection, these features were able to make ammonium tartrate the subject for more investigations over more than two decades, this ranked ammonium tartrate the second after alanine from the point of view of EPR dosimetry systems according to the number of studies [15-20]. Other relevant studies extended to other tartrate compounds, these compounds were derived from tartaric acid, and have some common features, however, ammonium tartrate still considered the best among them [21-24]. This study aims to evaluate how far ammonium tartrate can replace alanine in EPR radiation measurements.

2. Instruments, Materials, and Methods

2.1. Radiation source and Radiation dose measurements

Gamma irradiation was executed using a Cesium-137 gamma rays of model GB-150 which was fabricated by the Atomic Energy of Canada Limited on April 1970 with the initial activity of 1000 Ci. (3.7×10^{13} Bq). Air kerma (K_{air}) was measured and evaluated according to the international Atomic Energy Agency (IAEA) code of practice TRS-(381) [25]. The determination of (K_{air}) was performed using the secondary standard dosimetry system of the National Institute of standards (NIS) - Egypt, which was calibrated at the Bureau International des Poids et mesure (BIPM), France. Air kerma (Kair) values were evaluated with an associated expanded uncertainty of about 0.9 % at 95%, level of confidence (coverage factor = 2). Irradiation was executed at normal room conditions in a Perspex phantom irradiation capsules, range of radiation doses given to dosimeters was from 1.46 Gy to 2.5 kGy.

2.2. EPR system

The EPR spectrometer used in this study is an EMX-BRUKER EPR system, manufactured in Germany, which is supplied by a rectangular resonator 4102 ST cavity operating in the TE_{102} mode. The system is supplied with a 9.5 GHz microwave (X-band) Gunn-Oscillator Bridge.

2.3. Sample preparation and Evaluation method

Ammonium tartrate molecular formula is ($C_4H_{12}O_6N_2$), a molecular weight of 184.15 g/mol, and density of 1.6 g/cm3. The electron density <Z/A> for ammonium tartrate = 0.53217, Crystals of ammonium tartrate were purchased from ADWIC, prepared as described by Prolabo (99% for purity). Samples were prepared for irradiation by packing them in the irradiation capsules which were manufactured of leucite (Polymethyl methacrylate), (PMMA) in order to guarantee the equilibrium of charged particles during irradiation processes.

For EPR measurements of ammonium tartrate, parameters were as the following: microwave power was 0.6315 mW, modulation amplitude was 0.8 mT, 348.0 mT for the field center, 30.0 mT for the sweep width, 20.48 ms for time constant, and the conversion time was 10.24 ms for 1024 data points and hence the sweep time was about 10.48 s.

Empty tubes spectra were measured before recording samples spectra in order to assure the purity of the obtained EPR signals. A reference standard material (DPPH) was used for correcting the peak-to-peak amplitudes of the acquired EPR spectra where its EPR spectra were acquired before and after every single spectrum of ammonium tartrate dosimeters and hence eliminating all possible changes in the spectrometer sensitivity.

Masses of ammonium tartrate dosimeters were 0.20 ± 0.014 g, Normalization of EPR signals intensities was executed according to the mass of each dosimeter. The EPR spectrum of each dosimeter was recorded at least three successive times, each is of a single scan.

3. Results and discussion

3.1. Induced Radical:

Figure (1) represents the EPR spectra of ammonium tartrate dosimeters, where Figure 1A represents the unirradiated spectrum with no distinctive features and Figure 1B which represents a singlet located at g = 2.0049. This singlet is attributed to the radical: $H_4N^+ {}^-OOC\text{-}C^{\bullet}(OH)\text{-}CH(OH)\text{-}COO^-{}^+NH_4$ [16], while in [17] thoughts of another radical species has been started and there were several attempts to define the second stable radical in ammonium tartrate [20]. Both radicals share the same approximate position and hence it is difficult to resolve at room temperature, Figure 2 shows the EPR signal of irradiated ammonium tartrate recorded at modulation amplitude of 0.1 mT which confirms the presence of more than one overlapped singlets.

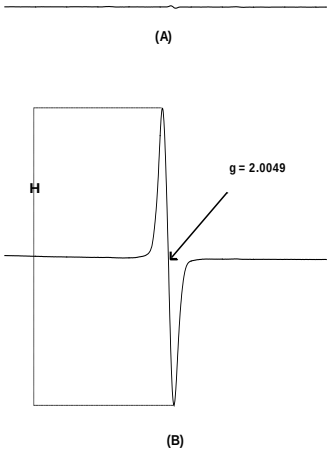

Figure 1. EPR spectra of ammonium tartrate, (A) unirradiated dosimeter, (B) 850 Gy gamma-irradiated dosimeter.

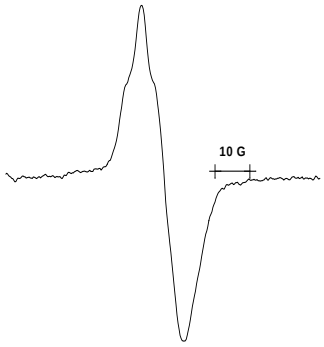

Figure 2. EPR spectrum of irradiated ammonium tartrate acquired at 0.1 mT modulation amplitude.

3.2. Time dependence

The time dependence curves of H_{PP} for both of the standard and ammonium tartrate dosimeters are shown in Figure 3, where it is clear that the instabilities of the peak-to-peak signal amplitude of ammonium tartrate over the first eight hours following irradiation cannot be attributed to the changes in the spectrometer sensitivity as can be confirmed by the behavior of the standard. During the first hour, variation in H_{PP} was in the range of 0.74% and the average value shows instabilities, while during the next 3 hours, H_{PP} decreased while the variation range was about 0.68%, after the 4th hour, H_{PP} started to increase apparently with variation range of 1.39%, this behavior is partially different from other previous studies [17, 20] and recommends the presence of more than one radical species.

In Figure 4, H_{PP} of ammonium tartrate was traced over 28 days following irradiation to 4 different doses, from the figure, H_{PP} increases till the day 2, however, variations over the first 3 days range was (0.41 - 0.89) %. At the end of the study term, H_{PP} showed a decrease to about 92% of its original value. In previous study [20], H_{PP} started to decrease only after the day 15.

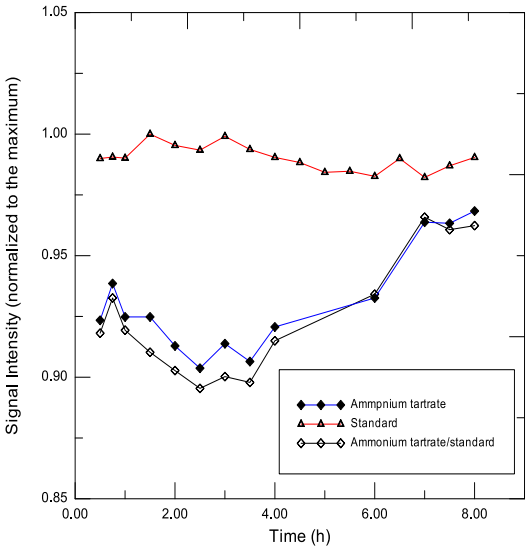

Figure 3. Short-term time dependence of Hpp over the first eight hours following irradiation.

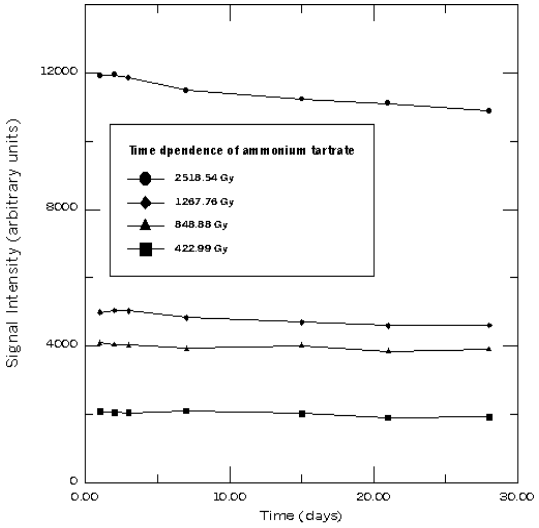

Figure 4. Long-term time stability of Hpp over the first 28 days following the day of irradiation.

3.3. Response to gamma radiation:

Figure (5), represents the response of ammonium tartrate and alanine dosimeters to the same radiation doses in the range (44-250) Gy and both were fitted linearly. From the Figure, it is clear that ammonium tartrate is more sensitive than alanine by a factor (on average) of about 2.1. The response to low radiation doses range (1.5-78) Gy is represented and linearly fitted in Figure (6), where ammonium tartrate dosimeters were found to be 1.84 more sensitive than alanine on average.

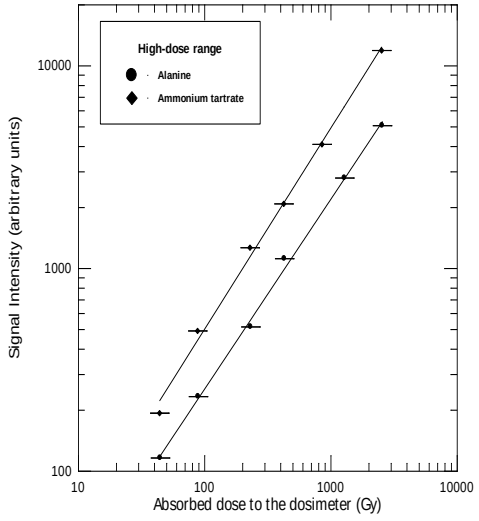

Figure 5. Response of Hpp for both of alanine and ammonium tartrate to radiation doses in the range (44-2500) Gy.

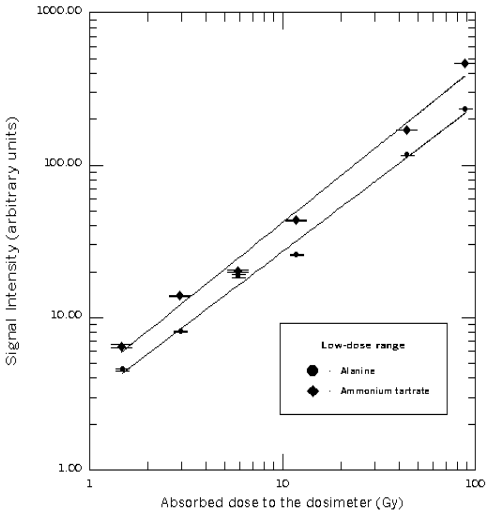

Figure 6. Response of Hpp for both of alanine and ammonium tartrate to radiation doses in the range (1.5-88) Gy.

Table 1 shows the percentage precision and the associated combined uncertainties for ammonium tartrate and alanine dosimeters for selected radiation doses over a wide range (0.57-2500) Gy. The table confirms the superior dosimetric features of ammonium

tartrate over the corresponding parameters of alanine, where ammonium tartrate shows better percentage resolution and lower uncertainties especially for low radiation doses.

Table 1. Percentage precession and the associated combined uncertainties for ammonium tartrate and alanine dosimeters for selected values of radiation doses.

Air kerma (Gy)	Ammonium Tartrate		Alanine	
	Percentage precision	Combined uncertainty	Percentage precision	Combined uncertainty
2500	0.06	0.48	0.14	0.48
1230	0.17	0.48	0.13	0.48
824	0.21	0.48	0.40	0.48
410	0.14	0.48	0.65	0.49
221	0.67	0.49	0.28	0.48
85	1.61	0.50	0.78	0.49
42	2.52	0.55	1.30	0.50
11	3.18	0.58	7.08	0.86
5.7	7.35	0.88	24.77	2.52
2.8	3.21	0.58	13.83	1.47
1.4	4.99	0.70	17.79	1.84
0.85	5.78	2.13	33.69	3.40
0.57	10.82	1.19	-	

Conclusion:

Ammonium tartrate dosimeters have common features with alanine, both are of complex EPR spectrum although the simple appearance of ammonium tartrate spectrum, both have complex time dependence, and on the other hand both of them possess tissue equivalency and possess linear response over a very wide range of radiation doses. However, ammonium tartrate showed more sensitivity toward radiation doses than alanine dosimeters, where their sensitivity is much better than alanine by a factor ranges from 1.84 to 2.1 times. Ammonium tartrate showed better percentage precision and lower values of associated combined uncertainties compared to alanine. From current study and previous studies also, ammonium tartrate can replace and can be used side-by-side with alanine in many of radiation dosimetry applications.

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