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ScienceDirect
Radiation Measurements 43 (2008) 1315–1318
Radiation Measurements
www.elsevier.com/locate/radmeas

Short communication

EPR of gamma-irradiated polycrystalline alanine-in-glass dosimeter

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Received 18 June 2007; received in revised form 18 March 2008; accepted 2 April 2008

Abstract

This study attempts to overcome some of the reported discrepancies in alanine-EPR reproducibility that may be related to alanine dosimeter preparation and/or EPR spectrometer settings. The dosimeters were prepared by packing pure polycrystalline L-α-alanine directly as supplied by the manufacturer in glass tubes. This dosimeter production scheme avoids any possible contribution to the EPR signal from a binding material. The dosimeters were irradiated with gamma ray to low-dose ranges typical for medical therapy (0–20 Gy). Special attention has been paid to the study of minimum detectable dose, measurement repeatability and reproducibility, and post-irradiation stability. The dosimeter exhibited a linear dose response in the dose range from 0.1 to 20 Gy. These positive properties favor the polycrystalline alanine-in-glass tube as a radiation dosimeter.

Keywords: EPR spectroscopy; Alanine dosimetry; Gamma radiation; Absorbed dose

1. Introduction

Alanine has gained world-wide acceptance as the standard material for radiation dosimetry using electron paramagnetic resonance (EPR) spectroscopy. When alanine is exposed to ionizing radiation, stable free radicals are created, and a resolved EPR spectrum with a low background appears. The intensity of the EPR signal from irradiated alanine is proportional to the concentration of radiation-induced radicals, which in turn is proportional to the absorbed dose. This dose response is highly stable, reproducible, cumulative, non-destructive, linear over 3–4 decades, and nearly independent to variations in dose rate, photon energy, radiation quality (low LET), minor temperature changes, and other ambient conditions (ISO/ASTM, 2004). These positive characteristics make EPR/alanine dosimetry a reliable technique for high-dose measurements in industrial radiation processes as well as medical irradiation processes, where lower doses are used (Desrosiers and Schauer, 2001; Schauer et al., 2007).

Gel, film, and pellet are three well-known forms of alanine dosimeters, commercially available for the measurements of radiation doses. Many types of binders were used to prepare these forms of alanine dosimeters, e.g., paraffin, acetate polyvinyl, polyethylene (PE), polyvinyl chloride (PVC), etc. However, the binding material may introduce a background signal due to radiation and/or thermal aging (Hayes, 2000, and references therein; Jahan and McKinny, 1999; Zhang et al., 1991; Morsy and Shwehdi, 2006). Because of the binding purpose of these materials, their expected background signal will definitely influence the EPR measurements of the irradiated alanine at different dose ranges. Surprisingly, not many studies have been published on the use of pure alanine without a binder in clinical dosimetry. Only recently, pure alanine was proven to be a reliable in vivo dosimetric method in teletherapy (Ciesielski et al., 2003) as well as in brachytherapy (Schultka et al., 2006); however, its accuracy can be improved by careful selection of measurement conditions.

This study proposes the use of pure polycrystalline alanine-in-glass tube design as a commercial dosimeter. This design overcomes discrepancies in the alanine-EPR reproducibility reported by many research groups (Hayes et al., 2000; Nagy

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doi:10.1016/j.radmeas.2008.04.088

et al., 2002) and will enhance the reported alanine lower limit of detection (LLD) without employing very complex procedures (Chen et al., 2002; Sharpe et al., 1996).

2. Experimental

2.1. Preparation of alanine dosimeters

High purity (> 99%) analar polycrystalline L-α-alanine was obtained from Fluka AG, Switzerland. Alanine was used as supplied by the manufacturer and lightly packed into identical glass tubes (Wilmad LabGlass, USA) having the same height of 40 mm and wall thickness of 0.5 mm but different inner diameters. Cotton was used to keep alanine in place. Some of the tube ends were left unsealed. Others were sealed either with epoxy or by a torch after degassing with the help of a vacuum line.

2.2. Irradiation and EPR measurements

Alanine dosimeters were irradiated in air and at room temperature to doses up to 20 Gy using a 130 Ci ^{137}Cs gamma irradiation device (Shepherd, model 78-10). The dose rate of the source was determined to be 0.215 Gy/h using the standards of NIST (National Institute of Standards and Technology) in the USA. The absorbed doses were verified and determined to be accurate using commercial thermoluminescent dosimeters (TLD) as a secondary standard. Moreover, the charge particle equilibrium in glass was assured during irradiation since the maximum electron range in the irradiated glass was estimated to be less than 0.2 mm.

The EPR spectra of irradiated alanine dosimeters were recorded at room temperature using X-band EPR spectrometer (JEOL, model JES-RE1X) operated at 9.5 GHz, equipped with a cylindrical microwave resonator cavity operating in a TE_{011} mode with a 100 kHz field modulation frequency. The EPR acquisition parameters that obtained highest signal-to-noise ratio were as follows: 10 mW for microwave power, 1.25 mT for amplitude modulation, a time constant of 100 ms, with 4096 channels for 20 mT width (center field 366 mT). These parameters compromise between signal-to-noise ratio and resolution. Spectra were collected and analyzed using EWWin EPR software (Scientific Software Services, USA). The dose response of an irradiated alanine dosimeter was assessed using the maximum peak-to-peak amplitude of the first-derivative EPR spectrum. An external standard reference (Mn^{2+}/MgO) was used to correct for sensitivity variations in the spectrometer response. The average amplitude of the second and fifth Mn^{2+} lines was used to normalize alanine signal amplitudes because of the overlapping of the third and fourth lines with alanine lines.

3. Results and discussion

3.1. Alanine dosimeter optimization

Identifying the characteristic features of the EPR spectrum of alanine dosimeter was difficult for low doses in the hundreds of mGy due to the inherent low signal-to-noise ratio. It is therefore essential to optimize the proposed dosimeter dimensions to obtain the highest sensitivity. At first, the influence of the internal diameter of the dosimeter on the quality factor Q of the microwave cavity was examined. The Q is given by

$$Q = \frac{2\pi(\text{energy stored})}{\text{energy dissipated per cycle}} = \frac{\nu_{\text{res}}}{\Delta\nu} \quad (1)$$

where ν_{res} is the resonance frequency of the cavity and $\Delta\nu$ is the width at half height of the resonance peak (Poole, 1983). The calculated values of Q indicate that the sensitivity of the EPR measurement increases gradually by varying the diameter from 1 to 3 mm and then degrades drastically by about 20% at 4 mm. The 3 mm alanine-in-glass dosimeter was then selected to investigate the dependence of signal intensity on alanine mass. The results indicated that the EPR signal intensity increases linearly with the alanine mass and then plateaus. Alanine mass of 110 mg was found to be optimal since the variation in signal intensity does not exceed 1.5% by altering this mass by 5%. As a result, the height of the alanine column in the tube corresponding to 110 mg reached the height of the active zone of the EPR microwave cavity. Moreover, the sealing and degassing of the alanine-in-glass dosimeters were tested. The signal intensity of the irradiated unsealed dosimeter is found to be 25% and 30% more than that of the sealed and degassed dosimeters, respectively.

Finally, the 3 mm dosimeter containing 110 mg alanine-in-glass was used to estimate the uncertainty in dose measurements. This uncertainty depends mainly on the reproducibility and repeatability of the alanine EPR signal measurements which, in turn, depend on many factors that include spectrometer stability and sample positioning inside the cavity. Both reproducibility and repeatability tests were carried out on five alanine dosimeters irradiated to doses of 1, 2, 4, 8, and 16 Gy. In the reproducibility test, each dosimeter was positioned in the cavity and then 10 successive measurements of the EPR signal intensity were taken. The percentage standard deviation at any dose was less than 3%. In the repeatability test, another 10 acquisitions were taken by removing the dosimeter from the cavity after being measured once then randomly put back and measured again. The percentage standard deviation due to sample repositioning and/or orientation was less than 5%. Thus, careful positioning and multiple scan acquisitions will reduce the overall uncertainty in dose measurements to ∼ 3%.

3.2. Lower limit of detection (LLD)

The analysis of the presented EPR spectral results in Fig. 1 indicated that the optimization of the EPR/alanine system certainly improved the detection of radiation-induced signals down to 0.1 Gy using pure polycrystalline alanine dosimeters with small number of scans and short scan time, and without any special material handling or mathematical treatment. The obtained dose response curve presents good linear behavior in the dose range 0.1–20 Gy with linear regression coefficient of 0.999 (Fig. 2). The deviation of dose response from the 'zero' indicates that the background signal or 'zero dose' clearly con-

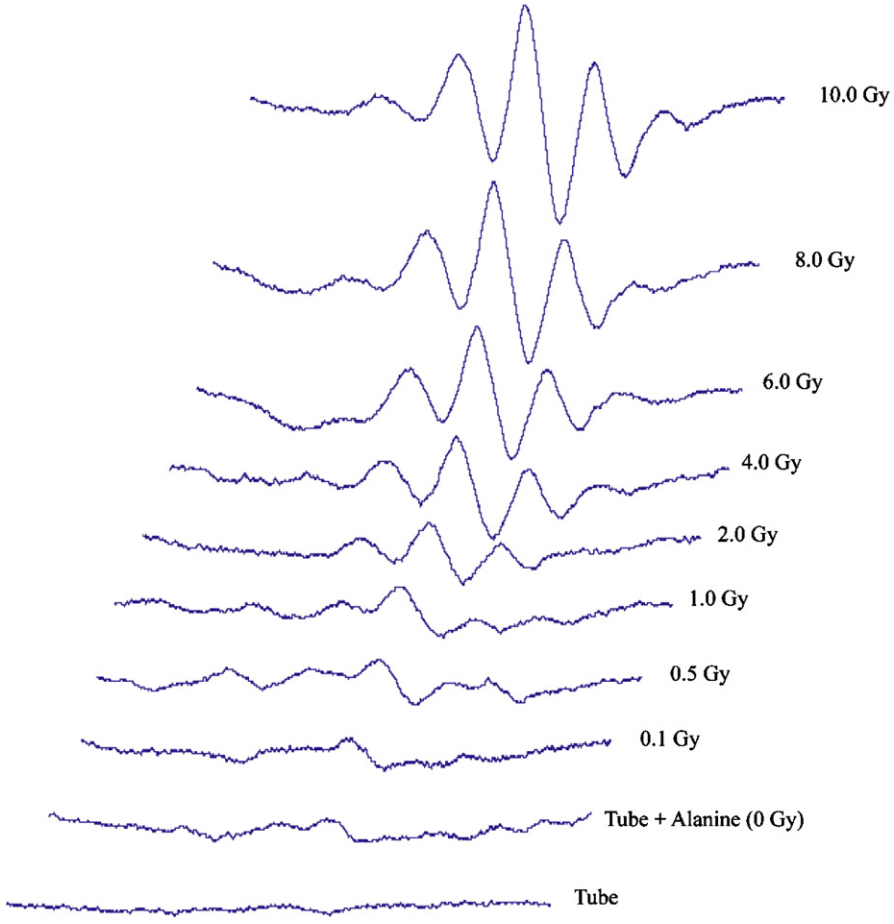

Fig. 1. EPR spectra of polycrystalline alanine dosimeters irradiated to different doses. The spectrum for the empty tube plus alanine at 0 Gy are also shown.

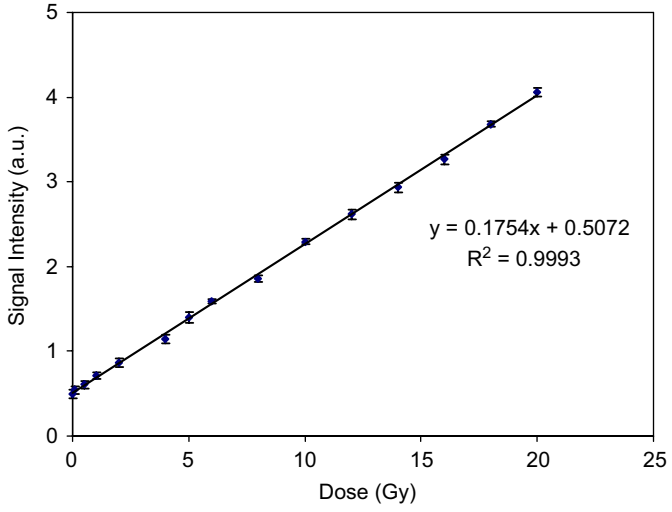

Fig. 2. Dose response curve for alanine dosimeters irradiated with gamma ray to doses in the range 0.1–20 Gy.

tains a measurable, non-radiogenic signal that is independent of dose and partly overlapped to the radiation-induced signal. This background has been attributed to endogenous paramagnetic impurities and/or preparation-induced defects (Wieser et al., 1993) and its value determines the LLD.

The LLD has been defined as the dose that produces an EPR signal amplitude significantly different from the background signal value by at least three standard deviations or higher (Bartolotta et al., 1993). With this definition and the data in Fig. 2, the calculated LLD for this EPR/alanine system is 0.3 Gy. Hence, there seems to be a large uncertainty in the measurement of the minimum dose (0.1 Gy) detected in this study due to the significant effect of background signal. This uncertainty at low doses is attributed to the combined effect of a low signal with instrument noise, variation in background signal (0 dose), and signal anisotropy (Nagy et al., 2002). In the previous studies, the lowest dose detection limit near 0.6 Gy was obtained (Sharpe et al., 1996). For extremely low doses, the usual procedure involves a complex parametric choice of EPR acquisition parameters and a low detection limit of 0.05 Gy has been achieved (Ruckerbauer et al., 1996).

3.3. EPR spectrum evolution with time

The EPR signal stability of the radiation-induced free radicals in gamma-irradiated polycrystalline alanine with time after irradiation was also studied. After irradiation all dosimeters were kept at room temperature in plastic bags and stored in the dark. The EPR spectrum of each dosimeter was recorded sev-

eral times during a period of one year after irradiation at the same spectrometer settings and the same ambient temperature and relative humidity conditions. The EPR measurements remained within 3% of the original measurement for the entire year. This result shows no signal fading upon long-time storage and proves the high stability of the polycrystalline alanine dosimeters.

4. Conclusions

A radiation dosimeter for EPR dosimetry technique using pure polycrystalline alanine-in-glass tubes was developed. This dosimeter production scheme avoids any possible contribution to the EPR signal from a binding material and increases the sensitivity of the dosimeter to low-dose gamma irradiation at optimum EPR recording conditions. Gamma-ray irradiation produced a sharp EPR signal at doses as low as 0.1 Gy. The dosimeter exhibited a linear dose response in the dose range from 0.1 to 20 Gy. The EPR signal was found unchanged in shape and intensity one year after irradiation. These favorable properties provided the potential of using polycrystalline alanine-in-glass as a commercial dosimeter for EPR dosimetry at low gamma irradiations doses.

Acknowledgments

The authors would like to acknowledge the support provided for this work by King Fahd University of Petroleum and Minerals, Dhahran, Saudi Arabia, through Fast Track project number FT-2004/18. The authors wish to thank M. Qureshi, N. Iqbal (Center for Applied Physical Sciences—Research Institute) and I. Ismail (Chemistry Department) for their valuable assistance.

References

Bartolotta, A., Fattibene, P., Onori, S., Pantaloni, M., Petetti, E., 1993. Sources of uncertainty in therapy level alanine dosimetry. Appl. Radiat. Isot. 44, 13–17.

Chen, F., Graeff, C.F.O., Baffa, O., 2002. Preliminary evaluation of second harmonic direct detection scheme for low-dose range in alanine/EPR dosimetry. Phys. Med. Biol. 47, 1357–1367.

Ciesielski, B., Schultka, K., Kobierska, A., Nowak, R., Peimel-Stuglik, Z., 2003. In vivo alanine/EPR dosimetry in daily clinical practice: a feasibility study. Int. J. Radiat. Oncol. Biol. Phys. 56, 899–905.

Desrosiers, M.F., Schauer, D.A., 2001. Electron paramagnetic resonance (EPR) biodosimetry. Nucl. Instrum. Methods B 184, 219–228.

Hayes, R.B., 2000. Concerns regarding recent NIST publications on alanine dosimetry. Comments Radiat. Phys. Chem. 59, 443–444.

Hayes, R.B., Haskell, E.H., Wieser, A., Romanyukha, A.A., Hardy, B.L., Barrus, J.K., 2000. Assessment of an alanine EPR dosimetry technique with enhanced precision and accuracy. Nucl. Instrum. Methods Phys. Res. A 440, 453–461.

ISO/ASTM 51607, 2004. Standard practice for use of an alanine-EPR dosimetry system. American Society for Testing and Materials.

Jahan, M.S., McKinny, K.S., 1999. Radiation-sterilization and subsequent oxidation of medical grade polyethylene: an ESR study. Nucl. Instrum. Methods Phys. Res. B 15, 207–212.

Morsy, M.A., Shwehdi, M.H., 2006. Electron spin resonance spectral study of PVC and XLPE insulation materials and their life time analysis. Spectrochim. Acta A 63, 624–630.

Nagy, V., Sholom, S.V., Chumak, V.V., Desrosiers, M.F., 2002. Uncertainties in alanine dosimetry in the therapeutic dose range. Appl. Radiat. Isot. 56, 917–929.

Poole Jr., C.P., 1983. Electron Spin Resonance, a Comprehensive Treatise on Experimental Techniques. Wiley-Interscience, New York, 125pp.

Ruckerbauer, F., Sprunck, M., Regulla, D.F., 1996. Numerical signal treatment for optimized alanine/ESR dosimetry in the therapy-level dose range. Appl. Radiat. Isot. 47, 1263–1268.

Schauer, D.A., Iwasaki, A., Romanyukha, A.A., Swartz, H.M., Onori, S., 2007. Electron paramagnetic resonance (EPR) in medical dosimetry. Radiat. Meas. 41, S117–S123.

Schultka, K., Ciesielski, B., Serkies, K., Sawicki, T., Tarnawska, Z., Jassem, J., 2006. EPR/alanine dosimetry in LDR brachytherapy: a feasibility study. Radiat. Prot. Dosim. 120, 171–175.

Sharpe, P.H.G., Rajendran, K., Sephton, J.P., 1996. Progress towards an alanine/ESR therapy level reference dosimetry service at NPL. Appl. Radiat. Isot. 47, 1171–1175.

Wieser, A., Lettau, C., Fill, U., Regulla, D.F., 1993. The influence of non-radiation induced ESR background signal from paraffin–alanine probes for dosimetry in the radiotherapy dose range. Appl. Radiat. Isot. 44, 59–65.

Zhang, W., Wang, H., Sun, J., 1991. Radiation effects on the blends of polyisobutylene. Fushe Yanjiu Yu Fushe Gongyi Xuebao. 9, 23–27.