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RESPONSE OF ALANINE DOSIMETER TO ULTRA-HIGH DOSE RATE ELECTRON BEAM

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Abstract. *The development of ultra-high dose rate (UHDR) platform machines and extensive studies on radiobiological effects have demonstrated a reduction in normal tissue toxicity via FLASH radiotherapy. Very high-energy electrons (VHEE) with energies ranging from 50 to 250 MeV have gained increasing interest to be employed as radiation sources for FLASH radiotherapy due to their ability to penetrate deeply seated targets. The delivery of high doses within sub-seconds (>40 Gy/s), pose significant dosimetric challenges. Conventional detectors suffer from saturation and ion recombination, leading to substantial errors and uncertainties in measurements. Alanine dosimeters can potentially be well suited for such UHDR beams. They are composed of organic crystalline amino acids. Alanine radiation characteristics are similar to those of tissue. Stable free radicals generated in irradiated alanine have unpaired electron which can be measured using electron paramagnetic resonance (EPR) spectrometers. The amplitude of the measured signal is correlated to the energy deposition i.e. dose. Alanine dosimeter is used as a secondary standard dosimeter in radiotherapy by several national metrology laboratories. Alanine is weak energy dependent within the therapeutic energy range (6-25 MeV). Its dose rate independence makes alanine a potential dosimeter for UHDR dosimetry. However, the response of alanine to very high energy electrons has not been reported, which is the chief aim of this research. Alanine pellets calibrated with Co-60 gamma-ray, were irradiated using 100 MeV electron beams from the Pulsed Energetic Electrons for Research (PEER) end station, which serves as the injector for the Australian Synchrotron. The linac can deliver electron pulses with pulse dose rate of 10^7 Gy/s. Six different dose per pulse (DPP) from 6 – 28 Gy per pulse (in single pulse of 200 ns time) were delivered to alanine pellets, with three pallets for each dose. The EPR spectra of irradiated alanine pallets were measured using Bruker EPR spectrometer. The amplitudes of the spectra were converted to absorbed dose to water using a calibration curve for alanine dosimeter irradiated with Co-60 gamma ray. The absorbed dose measurement of the alanine dosimeter irradiated with a 100 MeV VHEE beam is 16 % lower compared to the nominal dose as measured by Faraday cup. The relative response of alanine dosimeter for 100 MeV electron beam was 0.84. This result demonstrates the significant energy dependence of alanine dosimeters when exposed to a 100 MeV VHEE.*

Keywords: *alanine dosimeter, ultra-high dose rate electron beam, very high energy electron beam, FLASH radiotherapy*

1. Introduction

Recent studies have demonstrated that radiation delivered at ultra-high dose rates (> 40 Gy/s) within a sub-second time, known as FLASH radiotherapy, can reduce normal tissue toxicities while maintaining the same tumor response as conventional dose rates radiotherapy [1-4]. This differential biological response between the tumour and normal tissue, called the FLASH effect [1], has gained increasing interest as a promising approach for improving treatment outcomes in radiotherapy. Most FLASH radiotherapy and preclinical studies have been conducted with the electron beam. Favaudon et al. performed the recognized preclinical research on the FLASH effect using a Kinetron linac in 2014[1].

Currently, FLASH beams can be delivered in conventional (< 0.03 Gy/s) or ultra-high dose rate mode (electron pulse dose rate more than 2 x 10^7 Gy /s). Some clinical linacs have been modified to deliver electron beams at high dose rates. However, electron beams in the 4-20 MeV energy range have limited depth penetration, restricting their preclinical studies to small animals or superficial lesions [5, 6]. Very-high-energy electrons (VHEE) with beam energies ranging from 50 to 250 MeV have been proposed for dose delivery with greater penetration depth [7, 8]. Comparative studies of VHEE plans against clinical treatment plans (VMAT) indicate a reduction to organs at risk and an improvement in tumour conformity [9, 10]. The results make VHEE more attractive for FLASH radiotherapy.

Reliable and accurate dosimetry plays a crucial role in preclinical studies of FLASH radiotherapy. The standard protocols and equipment currently used are designed for conventional radiotherapy, with much lower dose rates (< 0.1 Gy/s). Several dosimeters have been used to measure dose delivery, but they have shown limitations when measuring dose in ultra-high dose rate beams [11]. Ionisation chamber is used as the standard dosimeters for absolute dose measurement in conventional radiotherapy. It has been demonstrated that existing recombination models and standard calculation protocols are insufficient for providing absolute dosimetry under ultra-high dose-rate beams

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[12, 13]. Many efforts have been made to apply to the ionization chamber response and develop a new ionization chamber for this purpose [14]. Diode detectors tend to exhibit over-response at high dose per pulse [15, 16]. Under an ultra-high dose rate pulsed electron beam, the diode detector exhibits saturation effects [17].

The Pulsed Energetic Electrons for Research (PEER) at ANSTO's Australian Synchrotron is currently developing a 100 MeV linear accelerator (linac) injector designed to deliver ultra-high dose rate electrons. The PEER Linac can deliver ultra-high dose-rate electron pulses, with a pulse dose rate of 10^7 Gy/s. PEER provides advanced research capabilities utilizing very high-energy electrons (VHEE) beams, a facility previously unavailable in Australia. Preliminary dosimetric investigations have been carried out at PEER employing a Moskin detector and scintillation screen, which identify a limitation associated with an in-vacuum fast current transformer (FCT) in relation to the charge delivery quality in air [18]. James et al. have successfully commissioned a Faraday cup to accurately measure the absolute in-air charge at PEER, serving as a means to verify the relative charge measurements between pulses during delivery—an essential requirement in dosimetry studies [19].

Alanine dosimeter is an organic crystalline amino acid ($C_3H_7NO_2$). Irradiation of this organic material generates stable free radicals in proportion to the absorbed dose. These free radicals, characterized by their unpaired electrons, can be measured using electron paramagnetic resonance (EPR) spectroscopy [20]. EPR is a technique used to study the properties of an unpaired electron in materials. Intrinsically, every electron has a magnetic moment and a spin quantum number with magnetic compositions $m_s = \pm \frac{1}{2}$, when an external magnetic field (B_0) is applied, the electron will align itself either parallel ($m_s = +\frac{1}{2}$) or anti- parallel ($m_s = -\frac{1}{2}$) to the magnetic field. Each alignment corresponds to specific energy due to Zeeman effect. The transition between the two energy levels is given by:

$$\Delta E = g_e\ u_B\ B_0 \qquad (1)$$

where g_e is the free-electron g factor equal to 2.0023, u_B is the Bohr magnetron and B_0 is external magnetic field.

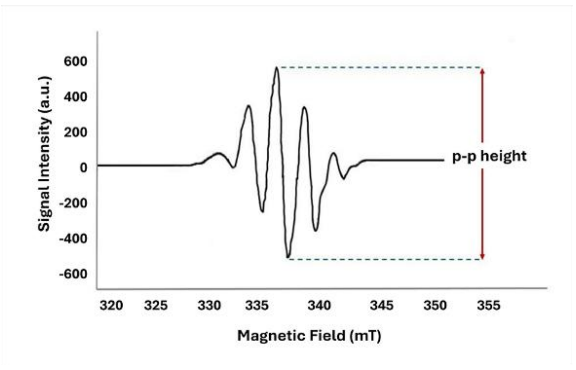

Figure 1. EPR spectrum of an alanine pellet irradiated with 50 Gy Co-60 gamma ray beam.

In EPR, free radicals are exposed to microwaves at a fixed frequency. By increasing the external magnetics field keeping the energy of microwave unchanged, the system is brought to the resonance condition. At this point, the unpaired electron can flip between two spin states. Typically, electrons are in the lower energy state, and the net absorption of energy is monitored and converted into a spectrum. The resulting alanine EPR spectrum consists of five peaks in a 1: 4: 6:4:1 ratio. In alanine/EPR dosimetry, the peak-to-peak amplitude of the central spectral line corresponds to the concentration of unpaired electrons generated in the alanine pellet, which is directly proportional to the absorbed radiation dose (Figure 1).

Alanine/EPR dosimetry has been utilized in industrial application such as food irradiation and medical device sterilization, especially in a high dose range (kGy) since the 1980s [21]. Key characteristics of alanine dosimeters include its stability of radical concentration, linear response over a wide range of radiation dose and non-destructive readout. Its near-water equivalent, effective atomic density (1.42 g/cm^3 and 6.8, respectively), makes it a weakly energy-dependent material. Alanine dosimeter is a dose rate independent detector that can be used at very high dose rates up to 3×10^{10} Gy/s using pulsed electron beams [22].

Currently, alanine dosimeters are used for photon and electron beams with energies ranging from 0.1 to 30 MeV (ISO/ASTM 51607) [23]. For the electron beam, the relative response of alanine compared with Co-60 irradiations is small [24-27]. Recently, McEwan et al. determined the consensus relative response of alanine to high-energy electron beam (6-22 MeV) based on Co-60 calibration from the overall published data to be 0.986 [28]. The alanine dosimeter has been used in the verification of dosimetry in electron FLASH radiotherapy, utilizing electrons up to 50 MeV [29]. However, the response of the alanine dosimeter to very high-energy electrons has not been studied.

This study aims to determine the dose response of alanine dosimeters to ultra-high dose rate electron beam relative to Co-60 gamma reference beam.

2. Materials and Methods

2.1. Alanine dosimeters

Alanine pallets (Far West Technology, Inc., USA, Lot number CP576) were employed in this study. Each pallet had a cylindrical shape with a diameter of 4.8 ± 0.01 mm and a thickness of 2.5 ± 0.1 mm. The nominal mass was 57.6 ± 0.02 mg, consisting of 90.9% L-alanine and 9.1 % paraffin wax binder by weight. The pellets were stored in a desiccator containing silica gel beads to minimize moisture.

2.2. Co-60 gamma irradiation

The Co-60 Gamma reference beam irradiation was performed with an Eldorado Co-60 teletherapy unit at the Australian Radiation Protection and Nuclear Safety Agency (ARPANSA). This procedure aimed to calibrate the electron paramagnetic resonance (EPR) signal of the alanine dosimeters to the absorbed dose to water. The absorbed dose rate was determined using a calibrated ionization chamber (PTW 30013, Serial No. 03587) positioned at a depth of 5 g/cm² with a field size of 10 × 10 cm² in a 30 cm × 30 cm × 30 cm water phantom. The calculated absorbed dose rate in water was 2.812 mGy/s, which was used to determine the irradiation time for dose delivery to the alanine dosimeter. Five alanine pellets arranged in a polymethylmethacrylate (PMMA) waterproofing sleeve NPL alanine holder were placed at the same position as the ionization chamber. The doses delivered to the alanine pellets were 2, 5, 10, 15, and 50 Gy, respectively.

2.3. Ultra-high dose rate electron beam irradiation

Alanine pellets were irradiated using very high-energy electrons of 100 MeV from the Pulsed Energetic Electrons for Research (PEER) end station at ANSTO's Australian Synchrotron. A 100 MeV electron LINAC serves as the injector for the synchrotron and can deliver ultra-high dose-rate electron pulses with a pulse dose rate of 10^7 Gy/s. Nine alanine pellets were inserted into holes in a Perspex phantom. The Perspex phantom, with dimensions of 10 × 10 cm², was drilled with nine holes, each 5 mm in diameter and 2.5 mm deep, to hold the alanine pellets. The spacing between the holes was 2 cm. The phantom was irradiated with a 1 cm build-up of a Perspex sheet. The EBT-XD film was inserted 3 cm behind the phantom surface to verify the beam position. During irradiation, the Perspex phantom was placed 255 mm from the 125 μm titanium exit foil window of the PEER LINAC. The Faraday cup, positioned 900 mm downstream, detected the absolute charge in air. Figure 2 shows the experiment setup.

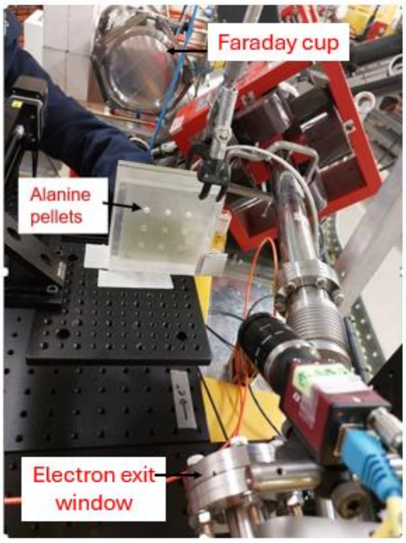

Figure 2. (a) Experimental setup of the Perspex phantom with alanine dosimeters at the PEER end station.

The charge delivered in each pulse was measured using the in-flange Bergoz FCT, sampled with a 14-bit ADC, and calibrated against a Faraday Cup at the start of each experiment. The expected peak nominal dose was calculated from the integrated pulse charge. The beam size and position stability were measured under the same conditions with EBT-XD. The estimated charge-to-dose conversion factor was 3.86 Gy/nC. Six different doses per pulse (DPP), ranging from 6 to 28 Gy per pulse (in a single pulse of 200 ns duration), were delivered to the alanine pellets, with three pellets for each dose or one dose in a row.

2.4. Electron Paramagnetic Resonance Spectroscopy

The signal from the irradiated alanine dosimeters was measured using a Magnettech MS-5000 tabletop electron paramagnetic resonance (EPR) spectrometer (Freiburg Instruments GmbH, Germany). Each dosimeter pellet was placed into a quartz sample tube with an inner diameter of 4.8 mm. To ensure precise positioning within the resonator, a polyethylene stand-off was inserted into the tube to hold the pellet at the centre. The parameters used for EPR signal acquisition are listed in Table 1. A calibration curve was established by correlating the EPR signal intensity with the absorbed dose to water, delivered using a Co-60 gamma reference beam.

Table 1. EPR signal acquisition

Parameters	Value
Microwave power	10 mw
Magnetic field range	320 mT – 360 mT
Modulation width	0.7 mT
Sweep time	60 s
Modulation frequency	100 kHz
Accumulation	4
Filter type	DIG

2.5. Relative response

The relative response (r_{Q,Q_0}) is the ratio of alanine/EPR response per unit absorbed dose for any beam quality (Q) to that for Co-60 gamma ray (Q_0) (equation 2). The relative response can be measured by the ratio of the slope of the calibration curve of the alanine dosimeter of given beam quality (S_Q) to the slope of the calibration curve with Co-60(S_{Q_0}) [25].

$$r_{Q,Q_0} = \frac{S_Q}{S_{Q_0}} \quad (2)$$

3. Results and discussions

Figure 3 shows the calibration curve of the EPR signal intensity of alanine dosimeters irradiated with Co-60 gamma rays and a 100 MeV electron beam. The calibration curves are fitted by linear regression. Both calibration curves show that EPR intensities increase linearly with the delivered dose. The correlation coefficients were 0.997 and 0.992 for Co-60 and 100 MeV electrons, respectively.

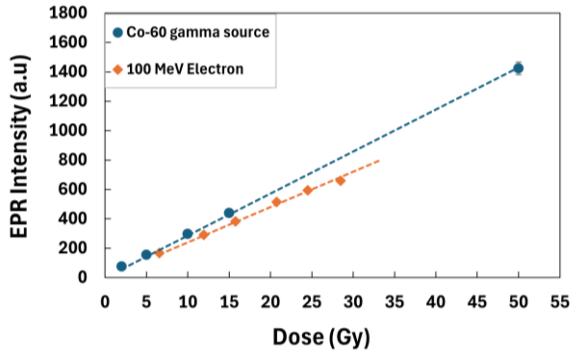

Figure 3. Calibration curve for Co-60 gamma rays and 100 MeV electrons.

The slope and standard deviation of calibration curves for a 100 MeV electron beam and Co-60 gamma ray were 24.0130 ± 0.0001 and 28.6150 ± 0.0001, respectively. The relative response of the alanine dosimeter of 100 MeV electrons relative to the Co-60 gamma ray, obtained by the slope ratio described in equation (2), is 0.84.

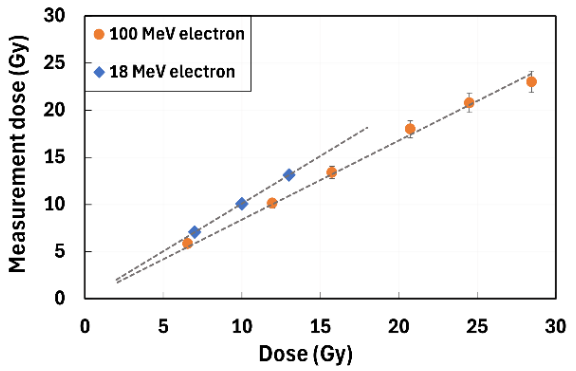

Figure 4. Alanine dose measurements from 100 MeV and 18 MeV electron beams.

Figure 4 shows the alanine dose measurement results compared with the delivered dose of a 100 MeV electron beam and an 18 MeV electron beam from a conventional linac. The curves illustrate the linear relationship between alanine dose measurements and the delivered dose, without energy correction. The slope or average ratio of measured doses to delivered doses for the 18 MeV electron beam is 1.01. Whereas, for the 100 MeV electron beam, the slope is smaller than that for the 18 MeV electron beam, representing a lowering of the alanine dose measurement results compared to the delivered dose, which is 0.84.

The study by Zeng et al. on the relative response of the alanine dosimeter in clinical electron beams ranging from 8 to 22 MeV to Co-60 is 0.987 [25]. They suggested that this difference in the alanine response may be interpreted as the gap between the alanine-to-water stopping power ratio for high-energy electrons and the alanine-to-water mass energy absorption ratio for Co-60. Changing bonding agents in alanine pellets did not change the energy dependence of alanine dosimeters as the study of Anton et al. confirms the hypothesis with a Monte Carlo simulation [26].

Considering the Bragg-Gray detector theory, alanine can function as a photon detector in a Co-60 beam and as an electron detector in electron beams. The relative response depends on the restricted mass collision stopping power ratio of alanine to water. For electrons, this ratio is lower than that of Co-60. However, it does not vary significantly across the electron beam energy range (6-22 MeV). It was noted that the detector geometry must be considered in the correction and that the field size may influence the change in radiation quality [26].

In our study, the relative response is 16% lower than that of Co-60. Figure 5 shows the comparison of doses delivered on alanine with doses measured from the gafchromic film (Gafchromic XD).

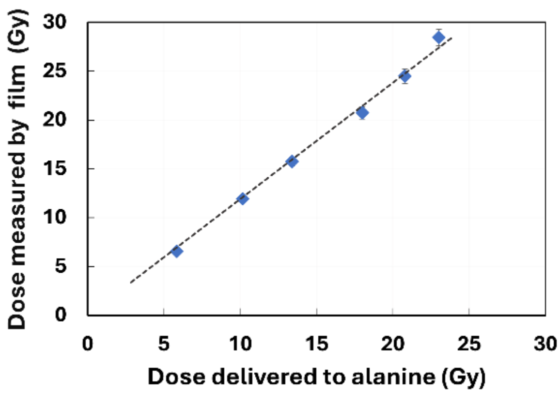

Figure 5. Comparison of delivered dose on alanine with doses measured with the gafchromic film.

Table 2. FWHM measurement

Dose (Gy)	X FWHM (mm)	Y FWHM (mm)
6.5	6.51	6.17
11.95	6.51	6.17
15.75	6.51	6.17
20.74	6.28	5.90
24.50	6.28	5.90
28.4	6.28	5.90

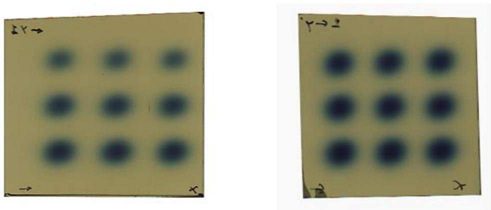

Figure 6. The irradiated films, produced using 100 MeV electrons with six different dose levels (one dose level per row), were used. From upper left to right: 6.5, 12, 15.7, 20.7,24.5, and 28.4 Gy.

As shown in the film (Figure 6), the 100 MeV electron beam has a FWHM of approximately 6.5 mm (x-axis) and 6.2 mm (y-axis), which is a small field (Table 2).

Small field dosimetry is challenging due to the loss of lateral charged particle equilibrium dose at a point, which is no longer equal to the energy deposited by charged particles entering and exiting the volume, as well as partial source occlusion or a mismatch between the detector and field size. Previous studies on the relative response of the alanine dosimeter in high-energy electron beams have been conducted under reference conditions (i.e., standard applicator, 10 × 10 cm, 100 cm SSD [24-27]. When using a 5 mm diameter alanine pellet to measure dose in a small radiation field, the detector size is comparable to the field size.

The alanine pellet measures the average dose over its volume. In a 6 mm field, the dose profile may exhibit steep lateral gradients, resulting in volume averaging and an underestimate of the peak dose, as well as reduced spatial accuracy. The volume averaging effect is primarily studied in small field dosimetry of photon energy, mainly due to treatment techniques like intensity-modulated radiotherapy (IMRT), volumetric modulated arc therapy (VMAT) and stereotactic radiosurgery using photon energy. The study of the volume averaging correction factor using alanine pellets for a 6 MV photon beam in a 1 × 1 cm^2 field size is 1.030. and less than 1.002 for the larger field sizes [30]. However, no investigation has been conducted into small-field electron beams. Therefore, the volume averaging effect of alanine dosimeters must be evaluated for small field electron dosimetry.

4. Conclusion

This study indicates that the relative response of the alanine dosimeter to an ultra-high dose rate electron beam at 100 MeV, in comparison to Co-60 gamma rays, was 0.84. This finding highlights the energy dependence of the alanine dosimeter in very high energy electron beam. Further research is necessary to address the volume averaging effects of the alanine dosimeter in small field dosimetry for electron beams.

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