

Investigation of Alanine Response to 100 MeV Proton Beam Irradiation

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1. Introduction

Alanine/Electron spin resonance(ESR) dosimetry has been established as a highly reliable method for quantifying radiation fields due to its excellent stability, near-tissue equivalence, and linear response over a wide range of absorbed doses. These characteristics make it a gold standard for both industrial processing and clinical applications, particularly in photon and proton beam experiments.[1,2] However, the vast majority of existing literature and characterization data are based on continuous or quasi-continuous irradiation conditions where the instantaneous dose rate remains relatively low. As modern accelerator technologies evolve, understanding how alanine behaves under extreme radiation environments—specifically those with high temporal peak intensities—is becoming increasingly essential for ensuring dosimetric accuracy.

The Korea Multi-purpose Accelerator Complex (KOMAC) utilizes a 100-MeV class proton accelerator that operates in a pulsed mode, delivering high peak dose rates during each pulse interval. Such unique beam characteristics present a potential challenge to standard dosimetry, as the high density of primary radicals generated during the pulse may influence the subsequent radiation-chemical reactions and radical recombination kinetics within the alanine matrix. In this work, we systematically investigate the response of alanine pellets under various pulsed beam parameters, including pulse frequencies and beam currents. By analyzing the ESR signal yields in relation to the total absorbed dose, this study aims to verify the linearity and robustness of alanine dosimetry in high-intensity pulsed proton fields, providing a critical validation for its use in advanced accelerator environments.

2. Methods

2.1. Sample Preparation and Irradiation

Commercial alanine pellets (FWT-50 series, Far West Technology, Inc.) were used for this study. Each pellet had a diameter of 4.8 mm, a thickness of 2.8 mm, and a mass of 58.2 ± 0.6 mg. For each irradiation set, four pellets were packaged together in a polyethylene bag. Irradiation was performed using a 100-MeV proton beam at the KOMAC. The samples were exposed to

cumulative doses of 1, 5, 10, 50, 100, 1000, 3000, and 5000 Gy. Post-irradiation, the samples were stored for one month at room temperature with a relative humidity maintained below 30%.

2.2. ESR Measurement

ESR measurements were conducted at room temperature. The magnetic field was swept from 3260 G to 3760 G with a sweep time of 30 seconds. To enhance the signal-to-noise ratio and ensure reproducibility, each sample was measured four times, and the resulting spectra were averaged for analysis.

2.3. Computational Modeling (DFT and EasySpin)

To elucidate the radical species and their ESR signal characteristics, density functional theory (DFT) calculations and EasySpin simulations were performed.

DFT Calculation: All DFT calculations were carried out using the Quantum ESPRESSO (v.7.2) package. The exchange-correlation interactions were treated with the Perdew-Burke-Ernzerhof (PBE) functional, employing standard pseudopotentials obtained from the official Quantum ESPRESSO distribution. The plane-wave cutoff energy was set to 240 Ry, and due to the large vacuum buffer (1 nm along each axis) surrounding the single alanine molecule, the Brillouin zone was sampled only at the Γ -point. After performing ionic relaxation on the pristine structure, the NH_2 group was removed to simulate the deamination process induced by proton irradiation. The resulting radical structure was further relaxed to determine its stable configuration. The GIPAW (Gauge-Including Projector Augmented Wave) method was then used to calculate the ESR parameters.

Signal Simulation: Using the EasySpin, we simulated the ESR spectra based on the calculated parameters to match the experimental results. This integrated approach allowed us to identify the origin of the observed signals and characterize the radical formation mechanism in the alanine matrix.

3. Results

Figures 1 and 2 illustrate the ESR measurement results for alanine pellets following irradiation with a 100-MeV proton beam. Although FWT-50 series

alanine pellets are commercially certified for a dose range of 100 Gy to 70 kGy, this study extended the investigation to a lower dose regime to evaluate the detector's sensitivity and linearity under pulsed proton fields. Consequently, the data were categorized into two sets: the low-dose range (1–100 Gy) and the high-dose range (1–5 kGy).

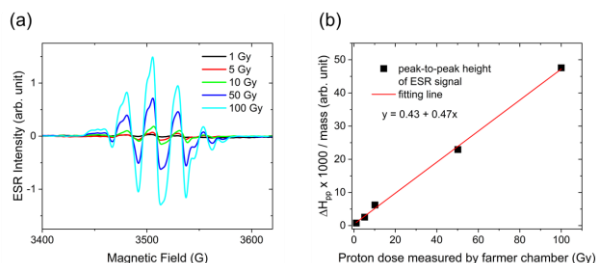


Figure 1. (a) ESR signals of alanine pellets with the low-dose range (1-100 Gy) (b) Proton dose rates vs the peak-to-peak amplitude of the central signal of ESR with mass-normalize

Figure 1(a) presents the representative ESR spectra. Within the magnetic field range of 3400 to 3600 G, a characteristic multi-line signal consisting of at least five distinct peaks was observed. This complex hyperfine structure is attributed to the stable radicals formed via the deamination of alanine molecules upon proton impact. As the cumulative dose increased, the overall ESR signal intensity exhibited a consistent and proportional enhancement, indicating a stable radical production rate even under high-intensity pulsed conditions.

To quantify the dosimetric response, the peak-to-peak amplitude (ΔH_{pp}) of the central signal was measured and normalized by the mass of each alanine pellet to account for minor gravitational variations. The resulting mass-normalized dose-response curve is shown in Figure 1(b). The data points were fitted using a linear regression model, which yielded an excellent linear correlation. The slope of the fitted line was determined to be 0.47, representing the sensitivity of the alanine/ESR system under the 100-MeV pulsed proton beam environment. This robust linearity, even at doses as low as 1 Gy (below the manufacturer's typical 100 Gy threshold), demonstrates the high reliability of alanine dosimetry for pulsed accelerator applications.

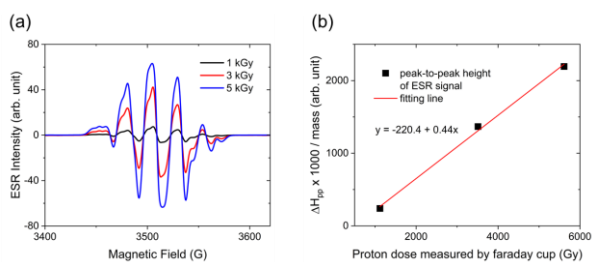


Figure 2. (a) ESR signals of alanine pellets with the high-dose range (1-5 kGy) (b) Proton dose rates vs the

peak-to-peak amplitude of the central signal of ESR with mass-normalize

Figure 2(a) shows the ESR spectra for alanine pellets in the high-dose range (1–5 kGy). The overall spectral shape is consistent with that observed in Figure 1(a); however, at the 5 kGy dose level, the shoulder signals became broader and their intensity increased to a level comparable with the main peaks.

The dose-response curve for this high-dose regime is presented in Figure 2(b), using the same normalization and fitting procedures as in the low-dose analysis. The slope of the linear fit was determined to be 0.44, which is similar to the value of 0.47 obtained in the low-dose range (Figure 1(b)).

Despite the similarity in slopes, a significant discrepancy was observed in the y-intercepts: 0.43 for the low-dose fit and -220.4 for the high-dose fit. This large difference in the intercept values indicates that the linear trends for the two dose regimes do not align on a single calibration line. This result suggests that using a single-point signal intensity for dose back-calculation across both ranges is currently challenging. Further detailed studies are required to establish more precise conditions for dose reconstruction in these pulsed proton fields.

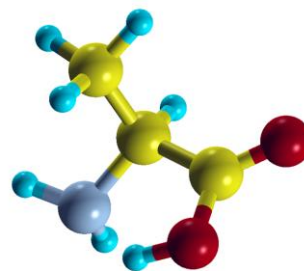


Figure 3. An Alanine structure

The ESR signal intensities before and after high-energy proton beam irradiation were compared using DFT simulations and the GIPAW method. It is well-established that exposure to high-energy radiation, such as gamma rays or proton beams, induces deamination in alanine, leading to the formation of stable free radicals that generate characteristic ESR signals.

Figure 3 illustrates the alanine structures used for the DFT calculations. In this study, the ESR signal intensities were evaluated by modulating the distance of the NH_2 group to simulate the radicalization process. The computational results showed that the signal intensity increased by more than 10^3 times following the simulated irradiation, rising from 1.58×10^9 in the pristine state to 2.69×10^{12} in the radical state.

To further investigate the origin of these signals, simulations were performed using the EasySpin program, as shown in Figure 4. The simulated spectrum that most closely matches the experimental data is presented in Figure 4(a). Additionally, Figure 4(b)

depicts the energy distribution and the Zeeman splitting as a function of magnetic field strength. The simulation confirms that the primary signals are distributed between 3400 G and 3760 G.

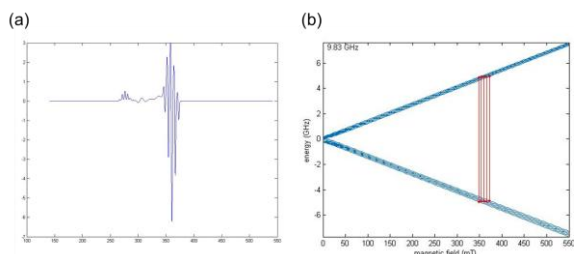


Figure 4. (a) Simulated ESR signal by EasySpin (b) Zeeman splitting for alanine structure

The simulated structure indicates that five hydrogen atoms contribute to the signal formation. As shown in the alanine model in Figure 3 (excluding the NH_2 group), there are exactly five hydrogen atoms within the radicalized alanine molecule, each appearing to act as a signal source. While the hydrogen atoms in a CH_3 group typically exhibit equivalent electronic distributions, the fact that five distinct hydrogen contributions were identified suggests the presence of an external factor influencing the electronic distribution. It is highly probable that the proximity or orientation of the NH_2 group (or its vacancy) induces this variation in electronic distribution, and we intend to conduct further research to clarify this mechanism.

4. Conclusion

In this study, we systematically evaluated the dosimetric response of alanine/ESR systems under 100-MeV pulsed proton beams at KOMAC. Our results confirmed a consistent linear response in both the low-dose (1–100 Gy) and high-dose (1–5 kGy) regimes, with comparable sensitivities (slopes) of 0.47 and 0.44, respectively. However, a significant discrepancy in the y-intercepts between these two ranges suggests that a unified calibration model for precise dose back-calculation remains a challenge under extreme pulsed conditions. Computational analysis through DFT and EasySpin further elucidated the radical formation mechanism, identifying five hydrogen-related signal sources and demonstrating a 10^3 -fold increase in ESR intensity post-irradiation. While this work validates the robustness of alanine as a reference dosimeter in high-intensity accelerator environments, further investigation into the electronic distribution effects and refined calibration protocols is necessary to enhance the accuracy of dose reconstruction across wide-ranging intensities.

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