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Quenching Behavior of Scintillation Light Yield in Ce-doped Gd_2SiO_5 under Proton and Alpha-particle Irradiation

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The scintillation light yield quenching of Ce-doped Gd_2SiO_5 (GSO) single crystals under proton and ^{241}Am α-particle irradiation has been investigated. Pulse height spectra of protons with the deposited energies of 1.5–3.4 MeV were measured at The Wakasa Wan Energy Research Center. In addition, pulse height spectra of ^{241}Am α-particles with the deposited energies of 1.62–5.41 MeV were measured. By comparing the experimental results of scintillation outputs with simulations, the Birks parameter of GSO was estimated to be 0.01 mm/keV.

1. Introduction

Scintillators are luminescent materials that absorb incident high-energy radiation and promptly convert it into low-energy photons.(1–3) These materials have widespread applications in fields such as medical,(4–6) security,(7–9) and environmental monitoring.(10–12) The required properties vary depending on the specific application; therefore, materials are selected on the basis of their physical and chemical properties, such as scintillation light yield, decay time, emission wavelength, density, effective atomic number, and hygroscopicity. New scintillators are continuously being proposed and developed with various material forms (e.g., crystal,(13–15) ceramic,(16–18) glass,(19–22) polymer,(23–25) and liquid(26–28)) to improve their performance and meet specific application requirements.

One such application in the medical field is heavy ion beam therapy,(29) a type of radiation therapy. Charged particles, such as protons and carbon ions, are accelerated to high energies by an accelerator and irradiated onto the treatment area of the human body. They lose energy through various processes. They interact primarily via Coulomb interactions with the orbital electrons of the absorbing medium, but also lose energy through Rutherford scattering and nuclear reactions with atomic nuclei, and via the emission of Cherenkov radiation and Bremsstrahlung. The energy loss per unit path length of the particles is known as the linear

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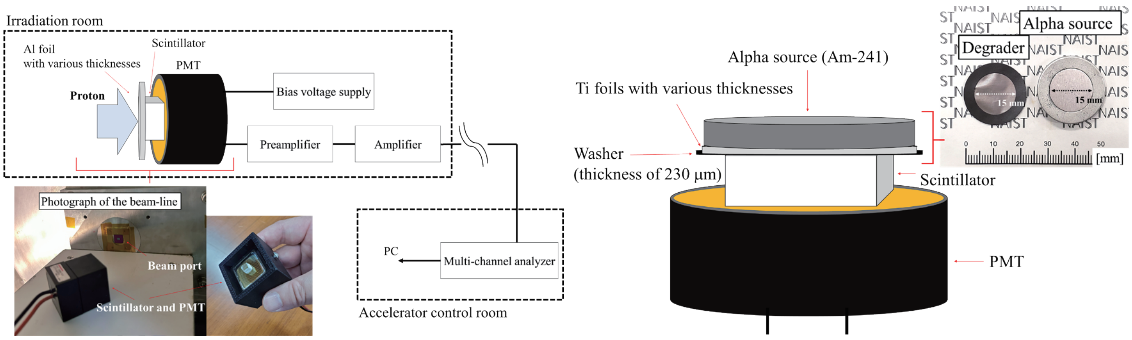
energy transfer (LET). The energy deposition distribution forms a large peak at the end of the particle's range. This peak is called the Bragg peak, and the peak becomes sharper as the charge of the particle increases. By aligning the Bragg peak with the target tumor, it is possible to deliver a high-energy dose to the target while depositing relatively low energy in the surrounding healthy tissue. This sharp dose gradient also means that even a slight deviation in the irradiation field can lead to significant underdosing of the target tumor and potentially cause severe complications by overdosing surrounding healthy tissues. Therefore, to enhance the safety and reliability of radiation therapy, it is essential to evaluate and monitor the dose in the vicinity of the tumor in real time during irradiation. While the application of storage phosphors,(30–33) such as those exhibiting thermoluminescence and optically stimulated luminescence,(34–38) has also been investigated, scintillators offer an advantage for such real-time applications.(39–41) However, for high-LET radiation such as heavy ions, the scintillation light yield shows a quenching effect,(42,43) which is often described by Birks' equation.(44) Consequently, it is essential to experimentally evaluate the light yield under heavy ion irradiation at various LETs. In this study, an experimental system for evaluating the scintillation light yield under heavy ion irradiation was tested by using commercial Ce-doped Gd_2SiO_5 (GSO, density:6.7 g/cm^3), whose scintillation temporal profiles at high LET we had previously investigated,(45) at The Wakasa Wan Energy Research Center. Furthermore, the quenching effects under α-particles were evaluated by using Ti foil degraders to evaluate their LET dependence.

2. Materials and Methods

0.5, 1, and 1.5% Ce-doped GSO (OXIDE, 5 × 5 × 5 mm^3 in size) scintillators were prepared for the following measurements. Pulse height measurements of protons from a tandem accelerator and ^{137}Cs γ-rays as a reference were conducted in irradiation room 1 at The Wakasa Wan Energy Research Center, Japan.(46) A schematic illustration is shown in Fig. 1(a). The scintillators were irradiated with protons in air using the same experimental setup as previously reported.(47) The beam window, which separates the vacuum and air, was made of silicon nitride with a thickness of 200 nm. The proton beam size was approximately 1 mm in diameter. The energies of protons were changed by inserting an Al foil (Nilaco) as a degrader between the beam port and the scintillator. The relationship between the thickness of the degraders and the deposited energy of protons is listed in Table 1. The energies were determined with a silicon surface detector (SSD). Here, the SSD was calibrated using ^{241}Am α-particles in air. Taking into account the energy attenuation in air and within the thin film on the active area of the alpha source, the energy of the ^{241}Am α-particles irradiating the SSD was determined to be 5.12 MeV. The calculation was conducted using SRIM.(48) The scintillators were combined with a photomultiplier tube (PMT, Hamamatsu Photonics, R7600U-200) using an optical grease (Shin-Etsu Chemical, KF-96H). High voltages (−600 V) were applied to the PMT using a bias voltage supply (ANSeeN, HV-01N). The PMT was connected to a preamplifier (ORTEC, 113) equipped with a resistor (150 kΩ), and the output signals were amplified and shaped by an amplifier (ORTEC, 572A). Subsequently, the obtained signals were transferred through a signal cable, several hundred meters in length, to a multi-channel analyzer (Amptek, Pocket MCA 8000A) connected to a control PC located in the accelerator control room (outside the irradiation room).



(a) (b)

Fig. 1. (Color online) Schematic illustrations of experimental setups for (a) proton and (b) ^{241}Am α-particle measurements.

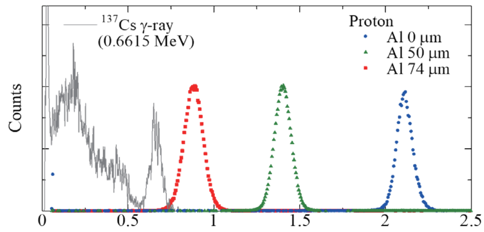
Table 1
Experimental conditions of energy degraders and energies of protons and ^{241}Am α-particles.

Degrader	Energy of proton (MeV)	Energy of α-particle (MeV)
None (Al 0 μm)	3.38	—
Al 50 μm	2.27	—
Al 74 μm	1.54	—
None (Ti 0 μm)	—	5.41
Ti 1 μm	—	5.18
Ti 2 μm	—	4.93
Ti 3 μm	—	4.69
Ti 5 μm	—	4.18
Ti 8 μm	—	3.37
Ti 10 μm	—	2.72
Ti 13 μm	—	1.62

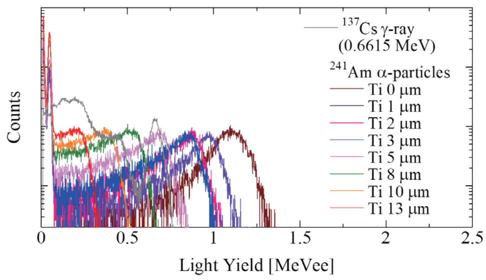
Pulse height measurements of ^{241}Am α-particles with various energies and ^{137}Cs γ-rays as a reference were conducted using a lab-made setup(49) equipped with a PMT (Hamamatsu Photonics, R7600U-200). To change the energies of ^{241}Am α-particles, degraders were prepared using washer-shaped Ti foils (Nilaco). The schematic illustration is shown in Fig. 1(b). The surface area of the Ti foil was adjusted to match the active area of the alpha source. Relationships between the thicknesses of the degraders and the α-particle energies are listed in Table 1.

3. Results and Discussion

Figure 2 shows the pulse height spectra of protons and ^{241}Am α-particles measured using the 0.5% Ce-doped GSO as a representative of the prepared Ce-doped GSO. The spectra of ^{137}Cs γ-rays were also measured to determine the channels of 0.6615 MeV as an electron equivalent light yield, which were unaffected by the quenching effect. Clear full-energy absorption peaks of protons and ^{241}Am α-particles, and photoabsorption peaks of ^{137}Cs γ-rays were observed. The peak channels of protons and ^{241}Am α-particles were compared with those of ^{137}Cs γ-rays, and the horizontal axes are displayed as the electron-equivalent light yields. As the deposited energies decreased, the light yields decreased. In addition, energy peaks were also observed at



(a)



(b)

Fig. 2. (Color online) Pulse height spectra of (a) protons and (b) ^{241}Am α-particles measured with a 0.5% Ce-doped GSO.

low-channel regions under ^{241}Am, whereas they did not appear under ^{137}Cs. It is noted that the peak positions did not change when the thickness of the degrader was changed. Therefore, the peaks originated from γ-rays emitted from ^{241}Am.

Figure 3 shows the relationships between the incident particle energy of protons and ^{241}Am α-particles and the electron-equivalent light yields of Ce-doped GSO. The error bars of electron-equivalent light yields were set to 10%, which was considered as systematic error. The scintillation light yield can be described by Birks' formula, which accounts for the quenching effect and is expressed as

$$\frac{dL}{dx} = \frac{S\dfrac{dE}{dx}}{1 + kB\dfrac{dE}{dx}}. \tag{1}$$

Here, dL/dx is the scintillation light yield per unit length, and S is the scintillation efficiency. dE/dx and kB are the deposited energies per unit length and the quenching coefficient, or Birks' parameter, respectively. dE/dx of protons at 1.50 and 3.25 MeV were respectively simulated to be 0.087 and 0.054 MeV/(mg/cm^2). dE/dx of α-particles at 3.00, 4.00, and 5.50 MeV were simulated to be 0.52, 0.45, and 0.37 MeV/(mg/cm^2), respectively. However, these energetic charged particles continuously lost energy as they traversed the material, and the dE/dx also changed continuously. To account for this process, the scintillation light yield was calculated using the user-defined subroutine "usrdfn2.f" in the Monte Carlo simulation code PHITS.[50] All samples showed a similar quenching trend as a function of incident particle energy in Fig. 3. The quenching

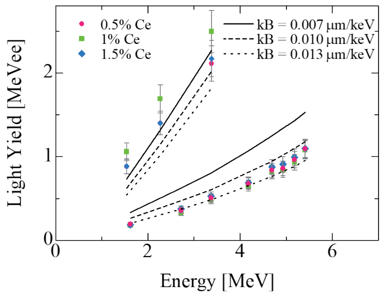


Fig. 3. (Color online) Relationships between light yields and energies of protons and ^{241}Am α-particles.

behavior is roughly characterized by *kB* values ranging between 0.007 and 0.013 mm/keV. Considering experimental uncertainties, such as the variability caused by inconsistencies in sample alignment, and synthesizing these results to derive a comprehensive *kB* value for GSO that is not specifically optimized for protons or α-particles, the overall quenching trend is considered to be approximated by *kB* = 0.01 mm/keV. The value was on the same order of magnitude as the value of GSO (0.017 mm/keV) under α-particle irradiations previously reported in past literature.(51,52) In addition, it was also on the same order of magnitude as the value of some other oxide crystals ($CdWO_4$: 0.013–0.027 mm/keV and $CaWO_4$: 0.010 mm/keV).(42)

4. Conclusions

We conducted pulse height measurements for proton and ^{241}Am α-particle with various energies using the Ce-doped GSO to evaluate its quenching coefficient. The deposited energies of protons were confirmed to be 1.54, 2.27, and 3.38 MeV, which were changed using Al foils. Clear full-energy absorption peaks of protons were observed, and the peak positions were shifted to lower channels as the deposited energies decreased. The deposited energies of ^{241}Am α-particles were observed to be 1.62, 2.72, 3.37, 4.18, 4.69, 4.93, 5.18, and 5.41 MeV, which were changed using Ti foils. Clear full-energy absorption peaks were observed, and the peak positions were shifted to lower channels as the deposited energies decreased. The quenching coefficient *kB* of Ce-doped GSO was estimated to be 0.01 mm/keV from the comparison of the experimental results of scintillation outputs with simulations.

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